

Local structural modification by femtosecond laser pulses and
light manipulation by dielectric metasurface hologram

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2025

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General Introduction

Light is one of the most fundamental phenomena in the nature. Humans can perceive the ever-changing scenes in the world mainly because our eyes can receive light emitted, reflected or transmitted by objects. Due to the close relationship between light and human life, optics, like mechanics and astronomy, is one of the earliest disciplines developed by humans. Since the very beginning of the human history, humans have tried to use light to light up the night, tell time with sundials and more.

In modern society, generations of researchers have discovered numerous methods to manipulate light. Among these, the laser (Light Amplification by Stimulated Emission of Radiation) stands out as one of the most significant inventions. The application of lasers remains a crucial area of study. Within the realm of laser development, the femtosecond laser ($1 \text{ fs} = 10^{-15} \text{ s}$) represents one of the most important breakthroughs in laser technology over the past century.

Femtosecond laser

Laser is a device that generates light through stimulated emission of radiation. It requires two key components: a laser cavity and a gain medium. The laser cavity confines photon propagation to a narrow frequency range and specific spatial direction, creating cavity modes. Laser radiation builds up in the cavity initially through spontaneous emission, followed by stimulated emission from the gain medium, where population inversion is established between laser levels. Continuous light amplification occurs via stimulated emission, and lasers can operate in continuous wave (CW) or pulsed modes [1], depending on their applications.

For femtosecond laser operations, pulsed laser techniques are crucial. First, to achieve nanosecond laser pulses, Q-switching (Q is the quality factor) is used. The

laser cavity may contain a Q-switch, which initially reduces the cavity and then gets switched to make the cavity Q higher after the lasing level of the gain medium is populated. Then, for picosecond pulses, mode locking is employed. Without frequency-selective elements, lasers oscillate on multiple resonator modes within the active medium's spectral gain profile, resulting in random fluctuations. By coupling the phases of these modes, coherent superposition is achieved, generating short picosecond pulses. Mode locking can be achieved by inserting elements into the cavity to lock the modes, resulting in a series of short pulses. This mode locking has been achieved by using optical modulators, by pumping the medium with another mode-locked laser (active mode-locking) or by saturable absorbers (passive mode-locking) or by a combined action of both locking techniques [2]. Synchronous pumping with a mode-locked pump laser ensures resonator length matching for ultrafast pulse generation. Conventional mode locking techniques typically produce picosecond pulses, while special techniques are required for femtosecond pulses.

The most commonly used techniques for generating high-power femtosecond laser pulses are colliding pulse mode-locking (CPM), Kerr lens mode-locking (KLM), and hybrid mode-locking [3]. Colliding pulse mode-locking (CPM) was the first technique to break the picosecond barrier. In this method, a CW ring dye laser is passively mode-locked using an absorber inside a ring resonator. Oppositely traveling short pulses collide in the absorber, doubling the total pulse intensity at the collision point. This increased intensity leads to greater saturation and reduced absorption, resulting in a net maximum gain. By carefully designing the amplifying gain and absorption losses, this mode of operation can produce short pulses as brief as 50 femtoseconds. A significant breakthrough was achieved with the discovery of self-mode-locking in a Ti:sapphire laser by de Spence et al [4]. This led to the development of an oscillator, a key component in many modern commercial laser systems now. In both active and passive locking methods, the nonlinear properties

of the amplifying medium are essential for the locking process. For self-locking modes, these nonlinear properties ensure that the modes can lock, either partially or fully, without the need for external modulation (active locking) or a saturable absorbing medium (passive locking). The amplifying medium must enable pulse narrowing with each round trip through the cavity. Kerr lens mode-locking (KLM) is the most widely used self-locking technique, utilizing the electronic Kerr effect to create an artificial fast saturable absorber [5].

For practical applications, high-power femtosecond lasers with tunable wavelengths should be applied. The rapid advancements in solid-state active materials, new nonlinear optical crystals, and mode-locking and amplification techniques have made ultrafast optical parametric amplifiers (OPAs: a phenomenon, for which a signal can be amplified using a parametric nonlinearity and a pump wave.) increasingly important. These OPAs are now a practical source of femtosecond pulses with tunable wavelengths across the visible and infrared spectral ranges [6]. OPAs are important tools for ultrafast spectroscopy, however high laser intensity is also necessary for strong field physics and more applications. Chirped pulse amplification (CPA) is a crucial concept for generating high-intensity, ultrashort laser pulses. The invention of CPA has significantly advanced Ti:sapphire laser technology, making the production of multi millijoule, ultrashort pulses a standard practice. The CPA process can lead to both amplitude and phase distortions, preventing the optimal recompression of the laser pulse and achieving high peak intensity. The process of stretching and recompressing pulses over many orders of magnitude in duration requires precise design and manufacturing of optical components, as well as careful construction of the stretcher and compressor [7]. Optical parametric chirped pulse amplification (OPCPA) combines the benefits of both optical parametric amplification (OPA) and chirped pulse amplification (CPA). This integration allows for the generation of high-intensity, ultrashort laser pulses

with tunable wavelengths, leveraging the strengths of both techniques. Such extreme high peak intensities of femtosecond laser pulses can greatly enhance the ability to investigate the laser-matter interaction, which will be described in Chapter 1.

Metasurface

One of the recent intriguing applications for femtosecond laser is the fabrication of metasurface [8-10].

Metasurfaces, ultrathin two-dimensional metamaterials with inhomogeneous sub-wavelength features, can provide an exciting platform for the development of ultrathin optics. When the light impinges onto the meta-atoms (nano-structures) of the metasurface, abrupt phase changes that can be controlled by the shape of the meta-atoms will be produced. Compare with the conventional optical components that manipulate the light by phase accumulation along the optical path under the restricted Snell's law, metasurfaces can modify the wavefront by introducing abrupt phase changes in the scale of sub-wavelength, called generalized Snell's law of refraction [11]. The schematic of the generalized Snell's law is shown in Fig. 1. For the normal Snell's law, the light propagates along the white dashed line, satisfies the equation:

$$n_t \sin \theta_t - n_i \sin \theta_i = 0 \quad (1)$$

where θ_t and θ_i are the angle of refracted light and incident light, n_t and n_i are the refractive indices of media on the transmission side and on the incident side. When a metasurface is introduced to the interface, just like a wall of linearly varying wall, the optical path of the light propagation will be different from the normal one as the yellow line following the generalized Snell's law [12]:

$$n_t \sin \theta_t - n_i \sin \theta_i = \frac{1}{k_0} \frac{d\varphi}{dx} \quad (2)$$

Where k_0 is the magnitude of the free space wavevector, $d\varphi/dx$ is the component of the phase gradient parallel to the plane of incidence.

Metasurface can be divided into two types: linear metasurface and nonlinear metasurface, according to the frequency change of the transmitted light [13]. Linear metasurfaces can manipulate the amplitude, phase profiles and polarization of the incident light and can be further divided by the working mechanism as: resonant phase metasurface, geometric phase metasurface, Huygens principle metasurface, transmission metasurface and so on [14-23]. However, in the nonlinear metasurface, light at new frequencies can be generated with the desired wavefront.

Thanks to the amazing capacity to manipulate electromagnetic wave within sub-wavelength scale, metasurfaces have attracted lots of interests in various fields, including wavefront shaping [24,25], polarization conversion [26-28], energy concentration [29,30], holography [30-33] and so on. Based on the applications on these fields, many meta-devices have also been demonstrated in the last ten years, such as meta-lens [34-36], absorber [37-39], invisible cloak [40,41] and so on. The dielectric metasurface hologram and some fabrication methods will be described in Chapter 1. It is very hard to fabricate ideal and precise metasurface. The unavoidable structural deformation always exists, such as rounded corners.

In this study, we focus on the structural modification by femtosecond laser direct writing and the light manipulation of dielectric metasurface hologram with rounded corners, including four chapters. The summaries of four chapters are shown below.

Summary of each chapter

Chapter 1

In this chapter, the theories of femtosecond laser-matter interaction inside transparent materials is introduced. Femtosecond laser-matter interaction inside transparent materials can be categorized into three regimes corresponding to the femtosecond laser intensity. Firstly, when the energy density in the interaction volume is below the ionization and structural damage threshold, plenty of effects are induced by the femtosecond laser in different materials, such as photorefractive effect, formation of color-centers, photo-darkening in chalcogenide glasses and so on. Secondly, at intermediate femtosecond laser intensity, the anisotropic nanostructures with birefringence, called nanograting, are generated inside glasses, crystals and so on. Finally, when the intensity of femtosecond laser is high enough, the plasma explosion happens, resulting in the cracks and voids. For the purpose of precise processing by femtosecond laser pulses, the size of the femtosecond laser induced region is also discussed. Generally, the size of femtosecond laser pulses inside transparent materials is around several hundred nanometers, due to the Airy disk and diffraction limit. However, in the case of nanograting, the width of the nanovoid can be as small as 20 nm and the period of the nanograting can be tuned from 140 nm to 320 nm. Femtosecond laser direct writing is also a highly potential fabrication method for metasurface, due to the highly precise modification on the surface of materials, as small as 20 nm. The characteristics and fabrication method of dielectric metasurface are also introduced. However, achieving the ideal and precise light manipulation by dielectric metasurface still remains challenging due to the deformation during fabrication, such as the rounded angles at the sharp corners.

Chapter 2

Femtosecond laser slicing is one of the key applications of femtosecond laser-matter interaction. Due to extremely high-power density and high spatial resolution, femtosecond laser slicing is an efficient and precise method for the cutting or slicing of hard-brittle materials, such as Aluminum nitride (AlN). AlN is one of the highly anticipated materials for deep-ultraviolet (UV) optoelectronic devices due to its wide-bandgap and high thermal conductivity. Growing a thick single-crystal AlN layer by Hydride Vapor Phase Epitaxy (HVPE) method on another AlN substrate generated by Physical Vapor Transport (PVT) method is an effective method to obtain pure single crystal AlN wafer. However, the waste of PVT-AlN is a serious obstruct for industrialization. In this chapter, the precise laser slicing of HVPE-AlN and PVT-AlN by femtosecond laser direct-writing technique is performed. The surface roughness of the separated HVPE-AlN is smaller than 120 μm , while the separated PVT-AlN substrate is successfully used to grow a new single crystal AlN by HVPE after polishing. Femtosecond laser double spots scanning and pre-modification by using a SLM are also investigated to improve the processing efficiency and precision.

Chapter 3

Femtosecond laser induced self-assembly periodic nanostructures (nanogratings) in SiO_2 glass have attracted enormous attention due to the form birefringence in the region of nanograting even though an isotropic material, i.e. glass. However, the physical mechanism of the nanograting formation is still not clear so far. In this chapter, we demonstrate the multi-layer nanogratings. The properties of the multi-layer nanogratings are strongly influenced by the polarization direction of the femtosecond laser for different layer. Besides, the birefringence behavior of the

nanograting has also an obvious change when controlling the distance between nanograting layers. Specially, when the distance between double nanograting layers is $5\text{ }\mu\text{m}$, where the depth of the first nanograting $Z_0 = 0$ and the depth of the second nanograting is deeper than the first one ($Z = 5\text{ }\mu\text{m}$), the double layer nanogratings perform as polarizers. When $Z < 5\text{ }\mu\text{m}$, the later nanograting overwrites the initial one. As for the case $Z > 5\text{ }\mu\text{m}$, the nanogratings exhibit birefringence independently. In the case of $Z < 0\text{ }\mu\text{m}$, where the second nanograting is written shallower than the first one, the properties of the nanogratings is quite different. When $Z < -15\text{ }\mu\text{m}$, the nanogratings exhibit birefringence independently. When $Z > -15\text{ }\mu\text{m}$, the later nanograting overwrites the initial one. When $Z = -15\text{ }\mu\text{m}$, the initial nanograting exhibit no birefringence while the second nanograting exhibit strong birefringence. This result shows that the top structure of the nanograting plays a most important roll for the birefringence of the whole nanograting.

Chapter 4

Femtosecond laser direct writing technique is considered as a highly potential method to fabricate micro/nano structures in various materials due to its high nanometer solution, precision, and large area processing. Recently, researchers use femtosecond laser direct writing to fabricate metasurface, which has attracted much attention in the last decade, due to its versatile capabilities to manipulate electromagnetic waves within subwavelength scale. Among the application of metasurface, dielectric metasurface hologram can significantly improve the resolution, efficiency, view angle and suppress the high order diffraction compared with other traditional devices. The ability to completely control the optical properties of light by metasurface is sensitive to the geometric parameters of the nanostructure. However, there still exists unavoidable structural deformation on the metasurface, which may strongly influence the functions of metasurface, by the current

fabrication methods, such as femtosecond laser direct writing technique. In this chapter, we introduce the rounded corners (fillets) to the sharp corners of the meta-atoms, such as rectangular pillars and isosceles triangular pillars, in dielectric metasurface and numerically study the effect of these fillets under different symmetric condition. We find that with some specific radius of the fillets, bound state in continuum (BIC) appears and makes the structure as an absorber. Except these BIC, the fillets, which don't break the original symmetry of the meta-atoms, have almost no effect to the manipulation of incident light by metasurface. However, for those fillets, which break the original symmetry of the meta-atoms, the amplitude and phase manipulation by metasurface may be distorted, especially for the isosceles triangular pillars.

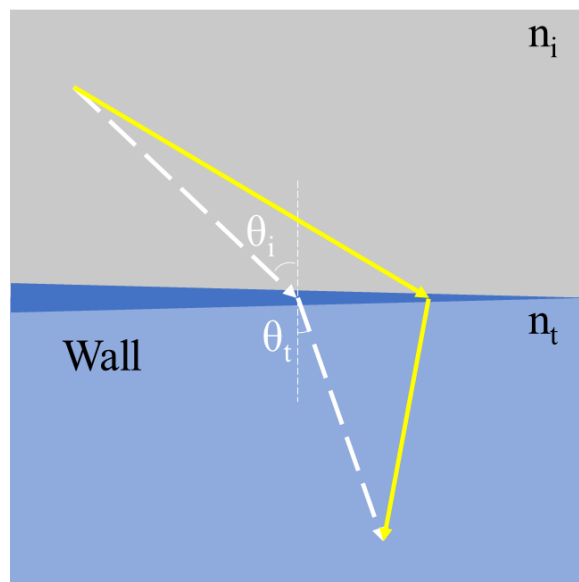


Fig. 1 The schematic of the generalized Snell's law of refraction.

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Chapter 1 Femtosecond laser-matter interaction and dielectric metasurface

1. Femtosecond laser-matter interaction

Laser is one of the greatest inventions in the humankind history and has enormously changed all aspects of our life. Among the laser development, femtosecond laser is one of the most important breakthroughs of the laser technologies in the last century and even the 2018 Nobel Prize in the physics was awarded to Professors Gerard Mourou and Donna Strickland for their groundbreaking work in chirped pulse amplification (CPA) technology which leads to an explosive improvement in the peak power of ultra-fast laser. Femtosecond (1fs = 10^{-15} s) laser is a pulsed laser with the pulse duration of femtosecond scale. The ultra-short pulse duration of femtosecond laser can create highly concentrated energy and realize an ultra-high peak power [1,2]. Such extreme high peak intensities in the focus of femtosecond laser pulses can greatly enhance the ability to investigate the laser-matter interaction [3-6]. One of the most promising application of femtosecond laser is the micromachining in the transparent materials, such as glass [7-9], crystal [10,11], polymer [12], and so on, due to the fact that intense nonlinear absorption happens when the femtosecond laser pulses with ultra-high peak energy are incident into the materials, promoting the valence electrons to conduction band [8,13]. The femtosecond lasers emit extremely short pulses, allowing for precise control over the interaction with materials. This precision minimizes thermal damage and enables the creation of intricate structures at the micro and nanoscale. Unlike traditional photolithographic processes, femtosecond laser fabrication does not require masks or complex setups, making it a more cost-effective solution for creating 3D structures.

The basic mechanisms of femtosecond laser interaction with matter has been extensively studied in the past two decades. While the exact mechanisms of femtosecond laser–matter interaction are not fully understood, the following scenario is widely accepted. When a single high-fluence femtosecond laser pulse irradiates into a transparent material, the laser energy is initially absorbed by the transparent material, generating photoelectrons. These electrons then transfer their kinetic energy to the lattice within a picosecond timescale. As a result, the heat diffuses, causing the material to melt and undergo fusion or explosion, leading to permanent structural changes. However, this is a qualitative description, and the specific phenomena induced depend on the chemical nature and physical properties of the materials as well as the laser irradiation conditions.

Typically, femtosecond laser–matter interaction is achieved by focusing the laser beam into transparent materials using a lens. The beam is then moved either transversally or longitudinally. The intensity distribution in the focal plane can be described by the well-known Airy disk. The minimum beam waist radius can be defined as the radius of the first Airy pattern:

$$r_{min} = 0.61 \times \frac{\lambda}{NA} \quad (1)$$

where λ is the wavelength of the femtosecond laser, NA is the numerical aperture of the focusing lens. The extension of the laser induced volume along the z -axis follows the Rayleigh length:

$$z_{min} = \frac{\pi r_{min}^2 n}{\lambda} \quad (2)$$

where n is the refractive index of the material. The focal volume can be reasonably defined as the region within the material where the intensity is equal to half of its maximum value when $r_{1/2} = r_{min}/\sqrt{2}$; $z_{1/2} = \pi r_{1/2}^2 n/\lambda$. Thus, the energy

deposition region of a femtosecond laser pulse inside the material can be simply calculated as [14]:

$$V_{1/2} = 2z_{1/2} \times \pi r_{1/2}^2 = \frac{0.683 \cdot n \lambda^3}{(NA)^4} \quad (3)$$

Then for example, in the condition at $n = 2.139$ (for AlN single crystal), $\lambda = 800 \text{ nm}$ (for Ti:sapphire femtosecond lasers), and $NA = 0.8$, one obtains $r_{1/2} = 0.43 \text{ }\mu\text{m}$, $S_{1/2} = \pi r_{1/2}^2 = 0.58 \text{ }\mu\text{m}^2$, $V_{min} = 1.83 \text{ }\mu\text{m}^3$. The minimum processing size of femtosecond laser direct writing here is around several hundred nanometers.

When the energy gap between the valence and conduction bands of the material is larger than the energy of a photon at 800 nm (commonly used with Ti:sapphire lasers), a single photon cannot induce an electron transition across the band gap, making linear absorption impossible. However, the extremely high peak intensity of the laser can result in nonlinear absorption, where an electron simultaneously absorbs energy from multiple photons, enabling the band gap transition [6,13]. The multiphoton absorption (MPA) rate ($P(I)$) strongly depends on the intensity of femtosecond laser (I) as described by the following power law:

$$P(I) = \sigma_k I^k \quad (4)$$

where σ_k is the MPA coefficient for the absorption of k photons.

The direct excitation of electrons by a strong femtosecond laser field is known as photoionization, which includes multiphoton ionization and tunneling ionization, depending on the laser frequency and intensity. When the femtosecond laser electric field is sufficiently strong, it significantly suppresses the Coulomb field that binds a valence electron to its parent atom. As a result, the bound electron can tunnel through the short barrier and become free. This process, known as tunneling ionization, plays a dominant role in femtosecond laser–matter interaction under strong laser fields and low laser frequencies. At high laser frequencies, multiphoton ionization dominates

the nonlinear femtosecond laser processing. Keldysh parameter describes the transition between the multiphoton ionization and tunneling ionization:

$$\gamma = \frac{\omega}{e} \left(\frac{mcn\varepsilon_0 E_g}{I} \right)^{\frac{1}{2}} \quad (5)$$

where γ is the laser frequency; I is the intensity at the focus; m and e are the mass and electron charge, respectively; c is the velocity of light; n is the refractive index of the material; ε_0 is the permittivity of free space; and E_g is the band-gap of the material. When the Keldysh parameter is larger than about 1.5, the multiphoton ionization dominates; otherwise, the tunneling ionization dominates.

Furthermore, an electron excited to the conduction band can sequentially absorb multiple laser photons, acting as a seed for a process called avalanche ionization. The energy absorbed by n electrons in the conduction band can surpass the band gap. In this case, the highly ionized electron releases excess energy to excite another electron to the conduction band through direct collision. The resulting two electrons in the conduction band can then repeat this process, ionizing more electrons. This leads to an exponential increase in free electrons. This ionization creates highly absorptive and dense plasma, facilitating energy transfer from the femtosecond laser pulses to the transparent materials. Consequently, many interesting phenomena can be observed during femtosecond laser processing.

As mentioned, numerous parameters influence the interaction between femtosecond lasers and transparent materials. These include the polarization (p), the laser light wavelength (λ), the pulse energy at the interaction point (E), the pulse density (D), the pulse duration (τ), the pulse repetition rate (f), the irradiation duration, the numerical aperture (NA), the material properties, and so on [8,14,15].

By considering the exposure parameters, material properties, and stages of material transformation, researchers have identified physical and chemical

modifications within various transparent materials induced by different femtosecond laser regimes by comparing the light intensity at the focal point [15-17]. There are basically three regimes of modification in transparent material: changes in optical properties by low intensity [18-20]; the anisotropic nano-structures with birefringence, called nanograting, induced by intermediate intensity [21-23]; the voids and cracks resulting from the high intensity caused plasma explosion [24-26].

When the intensity of femtosecond laser is below the optical breakdown threshold of materials, there are plenty of non-destructive effects on the optical properties, such as the photo-refractive effect [20,27], photodarkening in chalcogenide glasses [28,29], photo-polymerization of hybri materials [30,31] and so on. Specially, in glass, soft material transformations occur, resulting in refractive index contrasts ranging from 10^{-4} to 10^{-3} . In this case, the relaxation of electronic excitation does not generate sufficient heat to reach the material's softening threshold. Consequently, the thermomechanical effects seem negligible. Instead, laser-generated defects drive structural transitions, leading to denser packing within the matrix and resulting in changes to the refractive index [32-34].

By intermediate intensity, modifications induced by focused femtosecond laser radiation exhibit birefringence due to either laser-induced stress or the formation of self-assembled nanogratings with subwavelength periodicity, oriented perpendicularly to the laser light polarization, resulting in anisotropic reflection [33]. These modifications are enhanced by high incident energy intensity and slightly longer pulse durations, which provide stronger energy confinement and weaker plasma defocusing compared to shorter pulses. Thermomechanical effects such as pressure waves, compaction, cavitation, and rarefaction, along with signs of low viscosity regions, are evident in this trace. These effects lead to the formation, movement, and reshaping of voids into nanogratings [33]. The period of the nanogratings (Λ) is approximately following the equation below [7]:

$$\Lambda = \frac{2\pi}{\sqrt{\frac{1}{T_e} \left(\frac{m_e \omega^2}{3k_B} - \frac{e^2 N_e}{3\varepsilon_0 k_B} \right) - k_{ph}^2}} \quad (6)$$

where T_e is the electron temperature; m_e is the electron mass; ω is the angular frequency, k_B is the Boltzmann constant; N_e is the electron density; ε_0 is the permittivity of free space; k_{ph} is wave factor. The period of nanograting can be controlled from 140 nm to 320 nm. The width of the nanovoids in the nanograting is about 20 nm.

When the intensity of the femtosecond laser is above the optical breakdown threshold, the irreversible structural changes, often considered as damage, occur and can be used as a memory bit for high density 3D optical storage [35], the formation of optical waveguides [36] and in other photonics and material-processing applications [6]. With the high intensity of femtosecond laser pulses, a strong shock wave generated in the interaction region expands into the surrounding material and compresses it. This shock wave is followed by a decompressing rarefaction wave, which creates a void inside the material. The pressure at the shock wave front can reach several TPa, causing significant changes in the compressed material, forming a densified shell around the void. A key characteristic of a laser-driven micro explosion is that the modified material stays compressed and confined within a highly localized region inside the bulk material. A fitting formula was demonstrated by previous study [14] to describe the diameter of the void:

$$D_{void} = I_{abs} [A(E_{pulse} - E_{thr})]^{\frac{1}{3}} \quad (7)$$

where A is the absorption coefficient; I_{abs} is the absorption length; E_{pulse} and E_{thr} are the energy of pulses and threshold energy of material, respectively.

In this study, we focus on the generation of nanograting by intermediate intensity in chapter 3 and laser slicing in AlN single crystal by high intensity in chapter 2, respectively.

2. Dielectric metasurface hologram

Metasurfaces, made up of precisely engineered subwavelength units arranged in a two-dimensional plane, present a revolutionary approach to creating ultracompact optical elements and hold significant potential for the miniaturization of optical systems. One of the most attractive applications of metasurface is holography. Optical holography requires two steps. First, in the recording process, the phase and amplitude information of an object light is encoded as a hologram (interference fringes). Then, in the reconstructing process, the object light can be reconstructed by illuminating the hologram with the reference light. With the development of spatial light modulator (SLM) and computer, the hologram can be generated by numerical calculation, called computer-generated hologram (CGH) [37]. By CGH, it is possible to reconstruct arbitrary objects, greatly expanding the application of holographic technology. However, CGHs generated by SLM are suffering from the low resolution, low signal-to-noise ratios (SNR) and undesired high order diffraction, due to the limitation of pixel sizes of SLM, which are of the order of micrometers [38]. In recent years, plasmonic and high-index dielectric metasurfaces have been used to reconstruct the holograms with high-quality images. Since the meta-atoms of metasurfaces can reach the scale of sub-wavelength, metasurface holograms are able to achieve higher resolution, higher efficiency, wider view angle and suppress the high order diffraction in the far field, compared to the SLMs [39-42]. For example, a metasurface hologram made of a titanium dioxide nanoparticle-embedded-resin has achieved the conversion efficiency of 96.9% [42].

The methods and technologies developed for the fabrication of metasurface can be generally classified into four categories: direct-write lithography, pattern transfer lithography, hybrid patterning lithography and alternative techniques [43]. Among them, photolithography, a well-established technology in semiconductor manufacturing, also holds great promise for optical metasurface fabrication due to its high yield, large area, and mass production capabilities. In the traditional photolithography, light is used transfer a geometric pattern from a photomask to a light-sensitive chemical “photoresist” through a focusing lens. For the fabrication of metasurface with sub-wavelength features in several ten nanometers, shorter-wavelength light sources are preferred, which are known to cause damages to the lens and mask. To circumvent this technical difficulty, the maskless direct-write lithography has been developed which includes three different methods employing either particle beam, mechanics or laser to pattern nanostructures [44-48].

The femtosecond laser direct writing technique can also be used to fabricate metasurface [49-51]. Femtosecond laser is capable of emitting high-intensity pulses in a very short period of time, which can precisely ablate materials and form small heat-affected zones. Thus, high-resolution fabrication of tens of nanometers on transparent materials and semiconductor thin films have been demonstrated thanks to nonlinear absorption and localized light enhancement of femtosecond laser [52,53]. Femtosecond laser direct writing on the surface of the material begins with the creation of a nanohole, which serves as a seed. This nanohole is formed by a few femtosecond laser pulses that ablate the surface of the material through multiphoton absorption [54]. At the edge of the nanohole, the optical near fields are formed. The boundary condition requires that the normal component of the displacement (D) is continuous through the air-material boundary, following the equation below:

$$\varepsilon_a E_a = \varepsilon_m E_m \quad (7)$$

where ϵ_a and ϵ_m represent the permittivity in air and material. If the nanohole is small enough, the near fields at the opposite edges will interact and constructively enhance each other, with the enhancement being proportional to the square of the refractive index. The near field intensity enhancement inside the nanohole can be describe like [52]:

$$\kappa = (E_a/E_m)^2 = (\epsilon_m/\epsilon_a)^2 \quad (8)$$

and is proportional to n^4 . For the high-index material, this near field intensity enhancement is sufficient to carry out nano-ablation and confine the nano-ablation in the nanohole. The intensity around the nanohole is below the threshold required to cause ablation. Li et al. [52] fabricated nanostructures on titanium oxide with a sub-20-nm spatial resolution using this near filed intensity enhancement. Besides, Wang et al. [55] also used femtosecond laser direct writing technique to fabricate dielectric geometric phase optical elements with the structures of several hundred nanometers. Drevinskas et al. [56] demonstrated the potential implementations of ultrafast laser-induced periodic thin-film structures (nanogratings) as the geometric phase manipulating elements including polarization gratings, Fourier holograms, micro-lenses, and optical vortex micro-converters within the scale of several ten nanometers.

However, even the resolution of the femtosecond laser direct writing metasurface can achieve several tens nanometers, the unavoidable structural deformation always exists, such as rounded corners, which may strongly influence the functions of metasurface.

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Chapter 2 Femtosecond laser slicing on single crystal AlN layer grown by Hydride Vapor Phase Epitaxy on an AlN substrate generated by Physical Vapor Transport

1. Introduction

Due to the extremely high-power density and high spatial resolution, femtosecond laser slicing is one of the effective and precise methods for the cutting or slicing of hard-brittle materials, such as glass, Aluminum nitride (AlN) and so on [1].

Among the hard-brittle materials, AlN is a direct band gap semiconductor with a wide-bandgap (~ 6.0 eV) and a high thermal conductivity (~ 268 to 374 W/m $\cdot$ K) [2,3]. Thanks to these properties, AlN, especially the single-crystal AlN with low dislocation density, is one of the highly anticipated materials for deep-ultraviolet (UV) optoelectronic devices which have great potential for the application in high efficiency sterilization and virus inactivation [4-9]. For example, by mixed with Gallium Nitride (GaN), which has a bandgap of 3.4 eV (365 nm), the AlGaN can be generated and is expected to be used in deep-UV light emitting devices (the wavelength below 350 nm). Among these, especially, the development of $\text{Al}_x\text{Ga}_{1-x}\text{N}$ -based deep UV light-emitting diodes (LEDs) in the 260 nm band is now attracting much attention, as the absorption peak of the nucleic acids (DNA, RNA) is around 260 nm [10]. Traditional low-pressure mercury lamps (254 nm) have a large market, but come with drawbacks, such as containing mercury, short lifespan, high voltage operation, long startup time, lack of wavelength control, and low shock resistance. Therefore, if high-efficiency, long-lifespan deep UV LEDs can be realized, they could potentially replace low-pressure mercury lamps.

Due to the high melting point ($\sim 2200^{\circ}\text{C}$) of AlN, it is hard to grow bulk AlN single crystal from melt. The most extensively studied technique for growing bulk single AlN is the sublimation method, also called Physical Vapor Transport (PVT) method [5,11-14]. In this method, the high-purity AlN crystal powder is decomposed by being charged in a crucible at temperature above 2000°C and recrystallized at relatively lower temperature. This method allows for high-speed growth of AlN at rates exceeding $100\text{ }\mu\text{m/h}$, and crystals with extremely low dislocation densities ($<10^4\text{ cm}^{-2}$) can be obtained through spontaneous nucleation. However, due to the extremely high growth temperatures, atomic vacancies and impurity incorporation in the grown crystals are unavoidable. Consequently, while AlN single crystals grown by the PVT method have low dislocation densities, their deep UV light transmittance is reduced. Currently, when forming deep UV LED structures on PVT-grown AlN wafers, it is necessary to process the wafers to a thickness of about $20\text{ }\mu\text{m}$ after LED structure growth to reduce losses due to wafer absorption [15]. Therefore, it is necessary to elucidate the mechanisms that degrade the deep UV light transmittance of AlN single crystals and to achieve AlN wafers that combine low dislocation density with high deep UV light transmittance.

Another method that enables the bulk crystal growth of AlN is the Hydride Vapor Phase Epitaxy (HVPE) method [16-20]. This method grows crystals through the chemical reaction of chloride gases of group III elements and ammonia (NH_3) gas. It is the main method for mass-producing free-standing GaN substrates (wafers) necessary for blue-violet laser diodes [21-23]. However, compared to GaN HVPE growth, AlN HVPE growth has many limitations in terms of vapor phase raw materials and growth conditions. Recent studies [18-20] have demonstrated that the AlN HVPE growth can produce high-purity crystals with minimal impurity incorporation at growth rates of several tens of $\mu\text{m/h}$. On the other hand, an initial substrate is required for the growth by HVPE, and dislocation generation due to

lattice constant differences and cracking due to thermal expansion coefficient differences are major issues. There have been reports of obtaining free-standing AlN crystals by separating thick AlN films grown on (0001) sapphire or (0001) 6H-SiC wafers, but their dislocation densities were 10^7 - 10^8 cm⁻², which is insufficient for device fabrication. One solution is to use low-dislocation-density PVT-grown AlN wafers as the initial substrate for AlN HVPE growth, achieving low dislocation density through homoepitaxial growth and realizing the deep UV light transmittance characteristic of the VPE method.

In the case of grown a single crystal AlN by HVPE method on a PVT-grown AlN substrate, the HVPE-AlN was water-clear and free of stress and cracks, while the PVT-AlN substrate was amber in color with the presence of Al vacancies, substitutional impurities, and their complexes. In the previous study [20], the PVT-AlN substrate was removed by simply polishing, resulting in the waste of the substrates. In this section, the laser slicing of HVPE-AlN and PVT-AlN by femtosecond laser pulses is performed. In particular, the interaction between femtosecond laser and materials at the interface of two layers of AlN is revealed and the laser slicing by multiple spots is investigated. We also demonstrate that the PVT-AlN substrate after separation is able to be reused to grow a new HVPE-AlN.

2. Experiments

In the experiments, we use the samples provided by Tokuyama Corporation with thicknesses of 1 mm (400 μm for PVT-AlN and 600 μm for HVPE-AlN). The samples are cut into sizes of 5×5 mm and 2×5 mm. The experimental setup is shown in Fig. 1(a). We employ a regenerative amplified mode-locked Ti:sapphire femtosecond laser, centered at 800 nm with a pulse duration of 120 fs and a repetition of 1 kHz. The laser pulses are focused at the interface of the sample, from the HVPE side to the PVT side, using a 50 × objective (Nikon, NA=0.8). A spatial light modulator (LCOS-SLM, X10468-02, Hamamatsu Photonics K.K.) is utilized to compensate the spherical aberration and modulate the laser into multiple spots or other patterns. The SLM, sample stage, shutter, and laser pulse irradiation are controlled via sending commands from the serial port of a personal computer.

It is known that when a laser is irradiated into the material, which has a relatively high refractive index, with a deep irradiation depth, or the numerical aperture of the objective lens is high, the laser can't be focused properly. This is because of the spherical aberrations, which results in the laser modification region extending along the optical axis [24]. The AlN single crystal used in this study has a high refractive index of 2.139 ($\lambda=800\text{nm}$). Since precise three-dimensional focusing is essential for separating the two layers of AlN, an LCOS-SLM was used to correct these aberrations.

Fig. 2 shows the optical microscope images of the irradiation cracks observed from the side view, comparing the presence and absence of spherical aberration correction in AlN. Without the spherical aberration correction, the crack extends 186 μm , whereas with the spherical aberration correction, the extension is reduced to 104 μm , showing that the extension is suppressed obviously by the spherical aberration correction of SLM.

After the laser pulses are scanned across the entire interface of the sample, the sample is completely separated into two pieces using a Universal Testing System (Model 1122, Instron), which was shown in Fig. 1(b). A blade is placed perpendicular to the sample to facilitate the separation process. The surface roughness of the separated samples is measured by a surface profiler (Dektak150 BRUKER). Additionally, the separated samples are polished to expose the laser irradiation area. Energy-dispersive X-ray spectroscopy (EDS) measurements are performed from the top view of the irradiated area, while the electron probe microanalysis (EPMA) measurements are conducted from the side view of the irradiated area to evaluate the elemental distribution.

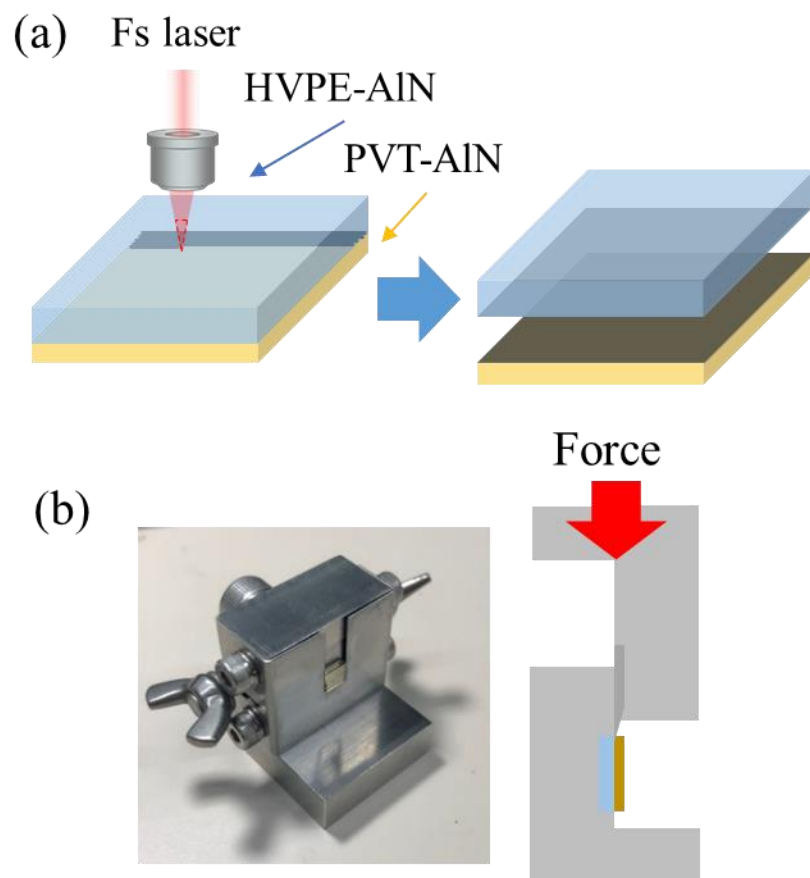
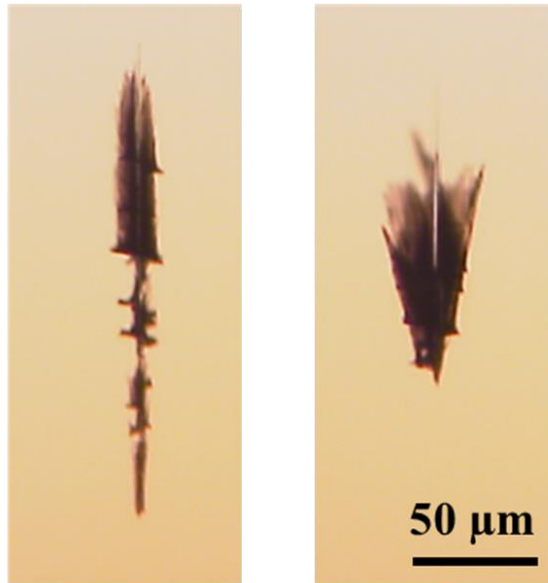


Fig. 1 (a) The schematic of the femtosecond laser scanning. (b) The equipment and schematic of the separation after scanning.



Without correction **With** correction

Fig. 2 The cracks caused by laser irradiation into the single crystal AlN, without and with the spherical aberration correction.

3. Results and discussions

3.1 Effect of the impurity concentration of AlN on crack formation

First, it is necessary to know the effect of the impurity concentration of the AlN sample on the crack formation. We focus the laser into four AlN samples with different impurity concentrations as shown in Fig. 3(a). The concentrations of Si, C and O are varying from 10^{16} atoms/cm⁻³ to 10^{19} atoms/cm⁻³. The pulse energy and the pulse number are 10 μ J and 1000 pulses, respectively. The results of the cracks formations are shown in Fig. 3(b). The optical microscope observations revealed that cracks are only formed in the samples with a Si impurity concentration lower than 10^{17} atoms/cm⁻³. One possible reason for crack formation when the Si impurity concentration is low is the increase in bond strength due to Si doping. It is known that Si substitutes for Al sites when doped into AlN crystals, and since the bond strength of Si-N (355 kJ/mol) is higher than that of Al-N (240 kJ/mol), it is considered that crack formation becomes hard in the case of high Si impurity concentration.

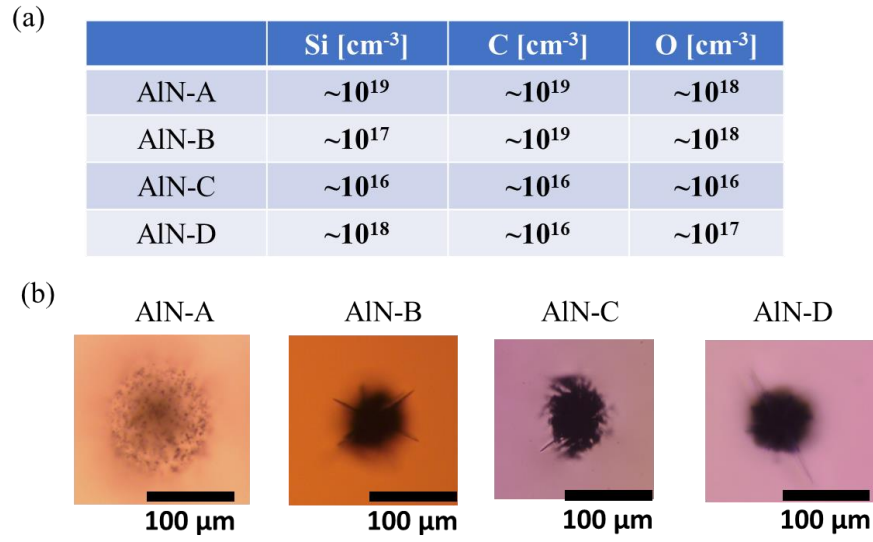


Fig. 3 The effect of the impurity concentrations of AlN to the crack formation. (a) The impurity concentrations of four different AlN samples. (b) The crack formation of the four AlN samples under the irradiation of femtosecond laser.

3.2 Laser damage threshold

It is known that the cross-section of multiphoton ionization in dielectric material is associated with the polarization state when irradiated by femtosecond laser, resulting in a polarization sensitive femtosecond laser damage threshold [25]. Here we measure the femtosecond laser damage threshold at different depth using circular polarized (CP) pulses and linear polarized (LP) pulses. Measuring the specific focus spot size at the sample interface is challenging, so here we use the single pulse energy to compare the behaviors of LP pulses and CP pulses. As shown in Fig. 4, under our experiment condition, due to the spherical aberration and nonlinear processing, the laser damage threshold increases with focusing depth, except at the interface. The lowest threshold is at the interface, primarily due to the unstable structure around the interface induced by the lattice mismatch of HVPE-AlN and PVT-AlN.

In addition, the threshold for LP pulses is consistently lower than that for CP pulses. This behavior can be attributed to the dominance of linear-polarization in the multiphoton ionization cross section when N is large, where N is the order of the multiphoton ionization [26]. The lowest femtosecond laser damage threshold is observed at the interface, which is about 580 nJ for CP pulses and 315 nJ for LP pulses. Based on our results, the LP pulse is chosen to be focused at the interface of the sample for the reduction of material cost.

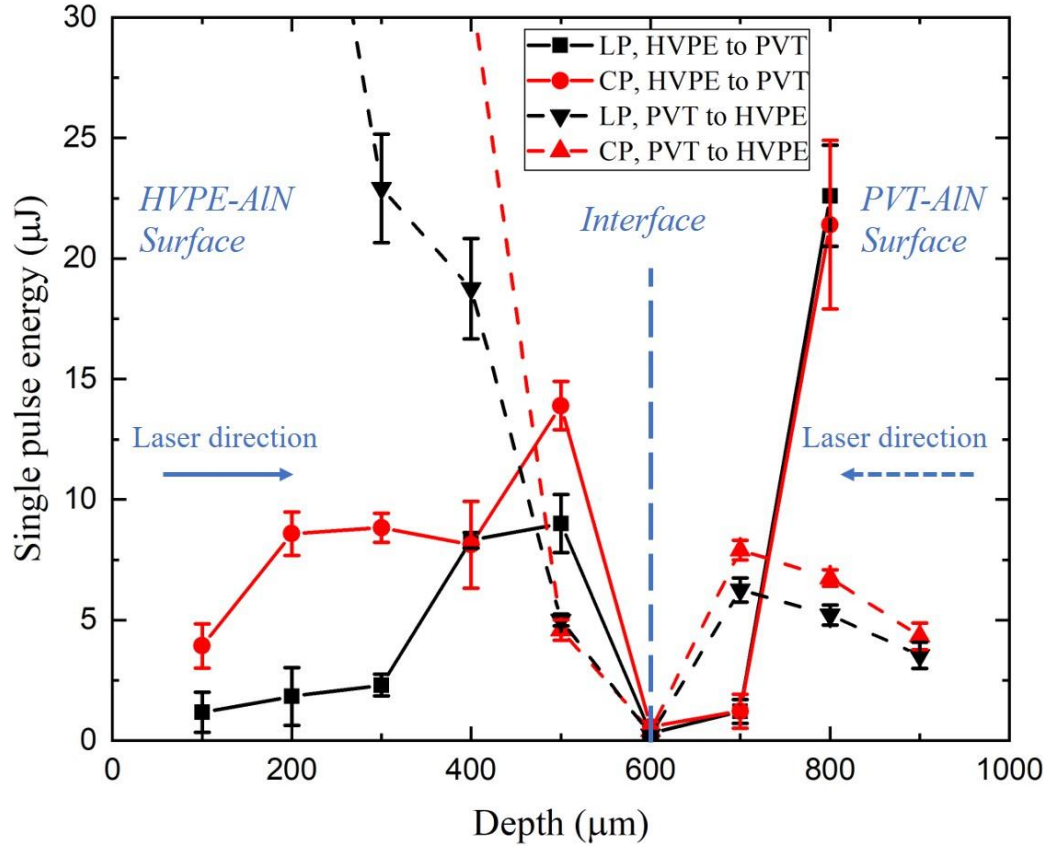


Fig. 4 The dependence of the laser damage threshold on depth for CP and LP pulses. The surfaces of HVPE-AlN and PVT-AlN are at 0 and 1000 μm respectively. The interface is at around 600 μm . The solid and dashed lines represent the laser irradiated direction from HVPE-AlN to PVT-AlN and from PVT-AlN to HVPE-AlN, respectively.

3.3 Parameters affecting femtosecond laser slicing

To optimize the laser slicing conditions, various experimental parameters are examined, including pulse energy, scanning speed, and laser polarization.

Pulse energy is a crucial factor in femtosecond laser-matter interaction as it determines the regime of laser modification [27]. For the purpose of laser slicing, the modification regime should be voids and cracks, which are resulting from the plasma explosion caused by high intensity [28]. Fig. 5(a) shows the scanning lines induced by femtosecond laser pulses at different pulse energies with a scanning speed of 100 $\mu\text{m/s}$. When the pulse energy is too low, the modification regime shifts to anisotropic nanostructure with birefringence [29], which do not provide sufficient damage for separation. Conversely, when the pulse energy is too high, cracks are generated randomly, making it difficult to maintain the evenness of the scanning lines. For our samples, the optimal pulse energy for femtosecond laser slicing is 10 μJ .

Another important parameter is the scanning speed. When the repetition rate of femtosecond laser is fixed, the scanning speed determines the pulse density, which indicates the energy accumulation per unit length. Fig. 5(b) shows the scanning lines induced by femtosecond laser pulses at different scanning speeds with a pulse energy of 10 μJ . A slower scanning speed causes more energy accumulation, resulting in random and stronger damage, making it hard to maintain a consistent scanning process. In stark contrast, when the scanning speed is very fast, around 2000 $\mu\text{m/s}$ equivalent to 0.5 pulses/ μm , the voids and cracks cannot be generated. Based on our results, a scanning speed at 1000 $\mu\text{m/s}$ is optimal for femtosecond laser slicing in our samples.

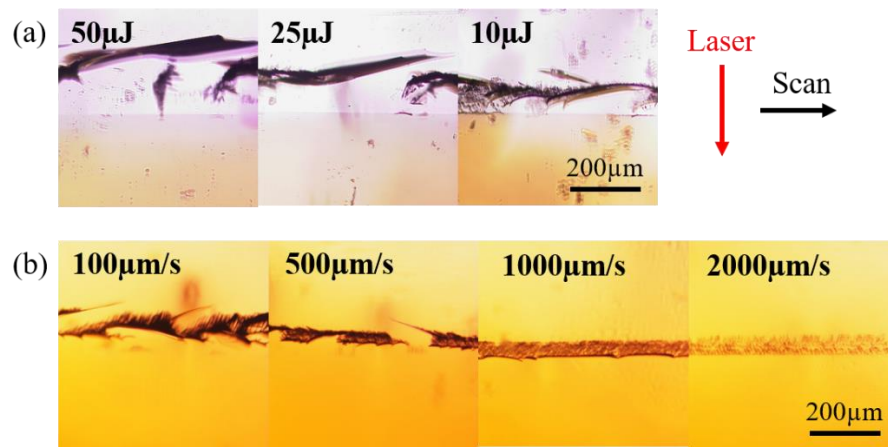


Fig. 5 The side view of the scanning lines induced by femtosecond laser pulses with (a) different pulse energy at the scanning speed of 100 $\mu\text{m/s}$ and (b) different scanning speed under the pulse energy of 10 μJ .

3.4 Separation after laser scanning

The laser pulses of 10 μJ are scanned with a pitch of 50 μm across the entire interface of the samples at the scanning speed of 1000 $\mu\text{m/s}$. The EDS mappings of nitrogen (N) and aluminum (Al) in the middle of scanning line, after polishing to the laser damage region, are shown in Fig. 6(a) and (b). The nitrogen element in the laser irradiated area decreases significantly, while the aluminum element increases. Fig. 6(c) and (d) show the SEM images of the unirradiated area and the irradiated area in Fig. 6(a) respectively. The fine uneven structures in Fig. 6(d) are caused by the explosion of N_2 during AlN is decomposed to Al (s) and N_2 (g) under the femtosecond laser irradiation.

After laser scanning, the sample is placed on the Universal Testing System, where a gradually increasing force is applied until it splits into two pieces. The necessary force to separate the sample is 530 N, as shown in Fig 7(a). The separated samples are displayed in Fig. 7(b), with HVPE-AlN on the left and PVT-AlN on the right. Fig. 8 shows that the average maximum height R_z , the sum of the maximum peak height and the maximum pit height, of three chosen lines on the separated HVPE-AlN is less than 120 μm , which is much smaller than other conventional method. In addition, the separated PVT-AlN is polished and used to grow a new single crystal AlN by HVPE. The X-ray topography (XRT) observation of the new HVPE-AlN is shown in Fig. 9. The contract in the image is due to the dislocations in the sample, primarily existing at the interface of PVT-AlN and HVPE-AlN. Compared to the previous studies [30], no apparent defect can be found in the image, providing the evidence of the successful growth of highly pure crystal.

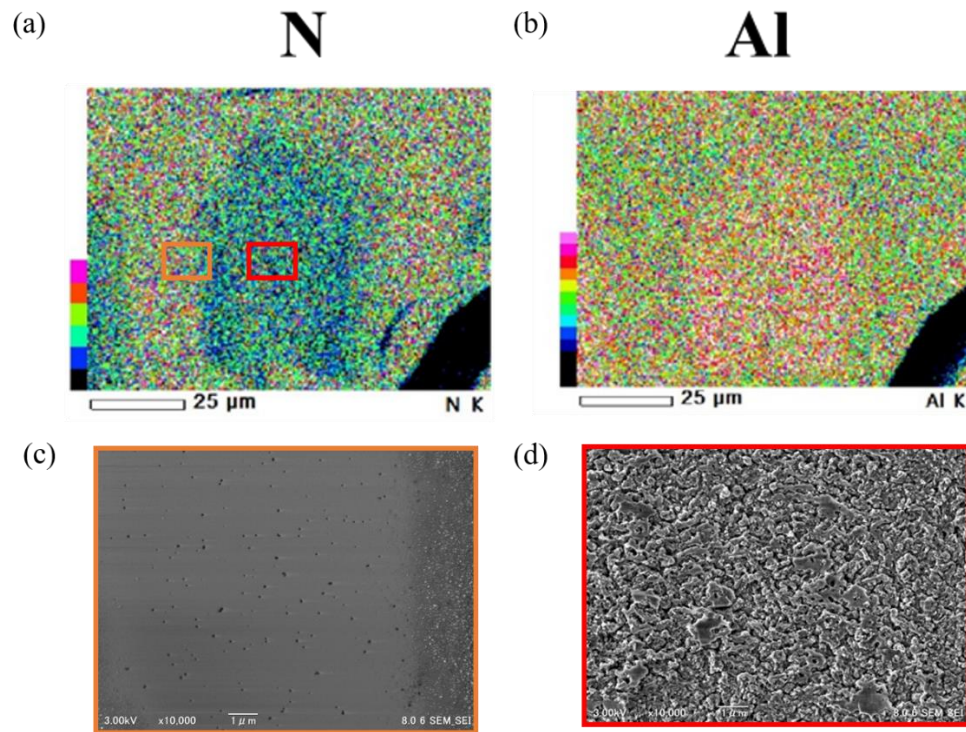


Fig. 6 The EDS mappings of N (a) and Al (b) in the laser irradiated area. The SEM image of (c) the orange rectangle area and (d) the red rectangle area in (a).

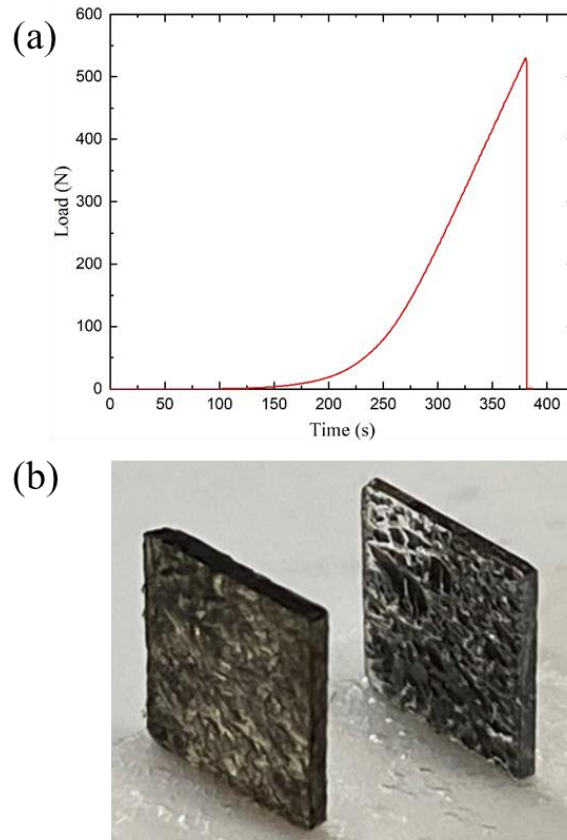


Fig. 7 (a) The force applied on the sample. (b) The completely separated sample, the left one was HVPE-AlN and the right one was PVT-AlN. (c) The surface roughness of HVPE-AlN after separation. (d) The XRT image of the new AlN crystal grown on the separated PVT-AlN after polishing by HVPE.

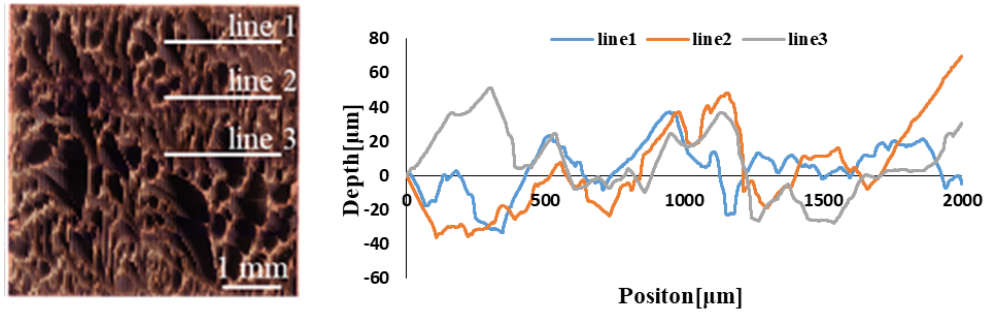


Fig. 8 The surface roughness of HVPE-AlN after separation.

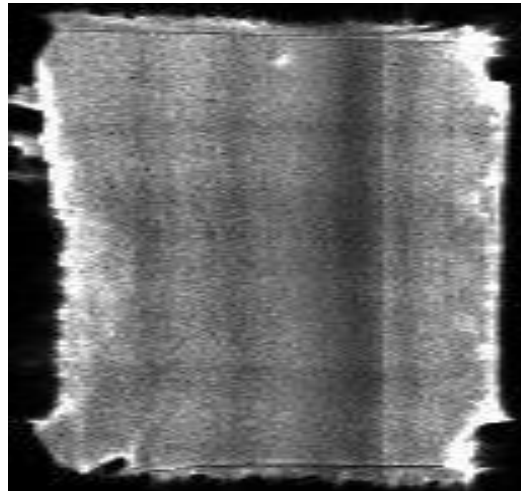


Fig. 9 The XRT image of the new AlN crystal grown on the separated PVT-AlN after polishing by HVPE.

3.5 Double spots femtosecond laser slicing

To enhance the efficiency of femtosecond laser slicing, we use a LCOS-SLM to split the laser focus into double spots. Previous studies have shown that the cracks generated by femtosecond laser multiple spots differ significantly from those generated by a single spot in some transparent materials. Additionally, simultaneous irradiation has a different effect on the crack generation compared to sequential irradiation [31]. In our sample, the cracks generated by femtosecond laser double spots are showed in Fig. 10, with a distance of 30 μm between the spots. When single crystal AlN is irradiated by a femtosecond laser from the c plane [0001], the cracks extend in the m-plane $[1\bar{1}00]$ and a-plane $[1\bar{1}20]$. However, the propagation velocities of the elastic wave in both directions, calculated based on the material parameters [32] are the same, indicating that the crack generation should be similar in both directions. Therefore, the most possible reason for the different lengths of crack A and crack B in Fig. 10(a) is the interaction between the two close spots. Fig. 11(a) shows the crack generation for simultaneous irradiation and sequential irradiation when the distance between the double spots is varied. Crack A is significantly elongated when the distance between the double spots is close to 30 μm under simultaneous irradiation, while it remains relatively unchanged under sequential irradiation. These results support our hypothesis well.

Based on the experimental results, we slice the samples using femtosecond laser double spots. After scanning the entire interface, the samples are separated using the same Universal Testing System. The forces applied on the samples are shown in Fig. 11(b). The necessary force for simultaneous irradiation is 627 N, while for sequential irradiation it is 794 N. Since simultaneous irradiation has a greater effect on crack elongation than sequential irradiation, the interface of the sample exposed to

simultaneous irradiation exhibits more cracks under the same laser pulse conditions and requires less force to be separated.

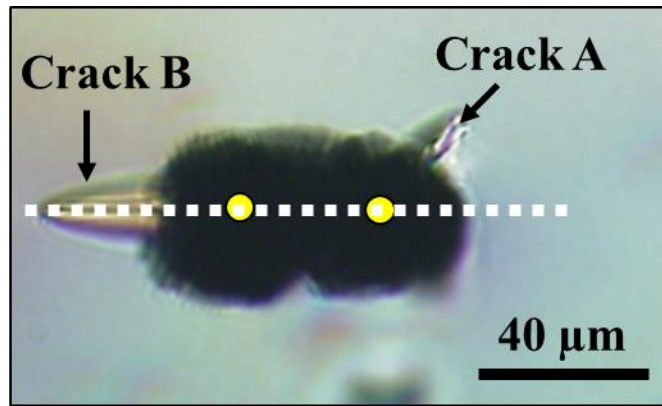


Fig. 10 The cracks generated by femtosecond laser double spots. (b) The lengths of the two kind of cracks as a function of the distance between the double spots by simultaneous irradiation and sequential irradiation. (c) The forces applied on the sample after double spots femtosecond laser slicing of simultaneous irradiation and sequential irradiation.

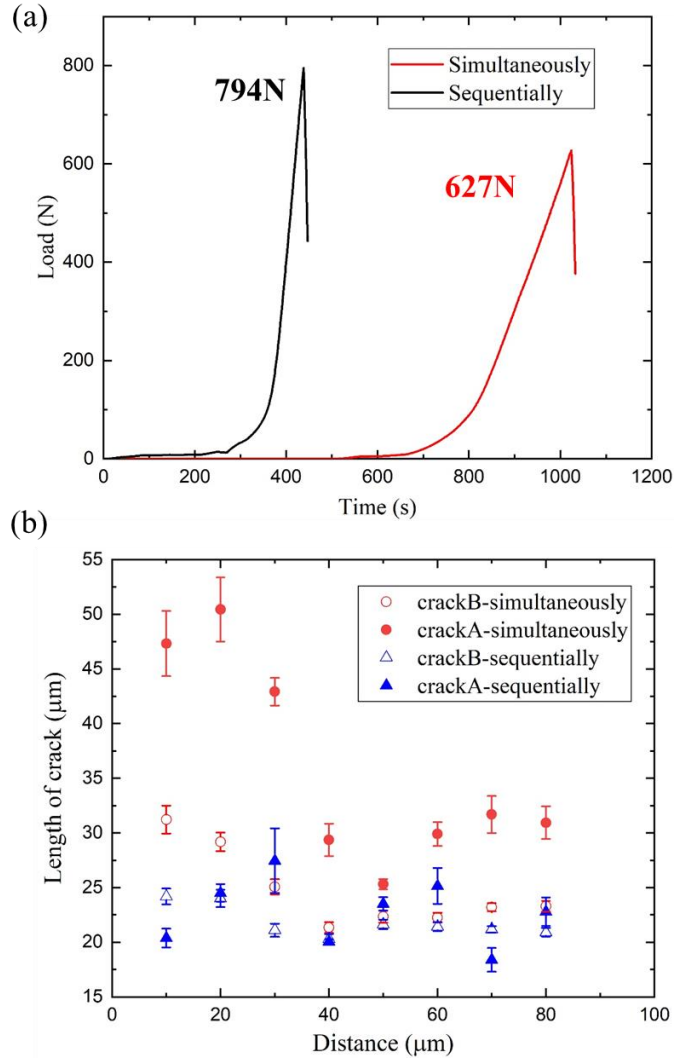


Fig. 11 (a) The lengths of the two kind of cracks as a function of the distance between the double spots by simultaneous irradiation and sequential irradiation. (b) The forces applied on the sample after double spots femtosecond laser slicing of simultaneous irradiation and sequential irradiation.

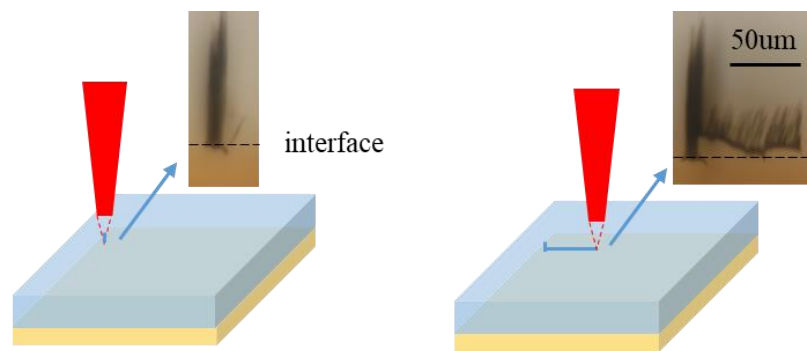
3.6 Pre-modification femtosecond laser slicing

We also employ the pre-modification method for femtosecond laser slicing on AlN. The schematic of pre-modification method is shown in Fig. 12, with insets displaying the side view of the laser damage regions. The left figure illustrates that a pre-modified laser damage region (pre-modified seed) is generated first and the right figure shows that the femtosecond laser is then scanned along the pre-modified seed.

The effect of the focus energy for pre-modified seeds, scanning energy for femtosecond laser scanning and scanning speed (related to the pulse number per micrometer) are controlled for pre-modification method. First, the side views of the scanning lines without pre-modification are shown in Fig. 13 (a). The scanning speed is fixed to 50 $\mu\text{m/s}$ while the pulse energy is changing from 60 μJ to 7.5 μJ . The results show that when the scanning energy is low as 7.5 μJ , the scanning line is hard to remain the continuity. However, from Fig. 13 (b), which shows the effect of the scanning energy, when the focus energy is fix to 7.5 μJ and the explosion time is 5s, to the scanning lines, it is known that with the pre-modified seed the scanning lines are much easier to remain the continuity, even the scanning energy is 1.5 μJ , and the damage of the scanning region is also much smaller than the normal methods.

Fig. 14 shows the side views of the scanning lines when the focus energy is fixed to 7.5 μJ and the scanning energy is fixed to 1.5 μJ while the scanning speed is changing from 100 $\mu\text{m/s}$ to 20 $\mu\text{m/s}$. The results show that when the scanning speed is too fast, which means the pulse number per micrometer is very small, it is hard to keep the continuity. When the scanning speed is too slow, the laser damage region will be enlarged obviously. The proper scanning speed in our experiment is about 50 $\mu\text{m/s}$.

Then the femtosecond laser is scanning over the whole sample with the focus energy of $7.5\ \mu\text{J}$, the scanning energy of $1.5\ \mu\text{J}$ and the scanning speed of $50\ \mu\text{m/s}$. After scanning, the samples are pasted to two pillars and can be easily separated by hand. The roughness of the samples is measured to compare with the normal methods. Fig. 15 shows the roughness of the samples measured by the Stylus Profilometer. The average maximum height R_z , the sum of the maximum peak height and the maximum pit height, of five chosen lines on the separated sample, is about $32\ \mu\text{m}$, which is much smaller than the normal methods.



Step 1: Generation of pre-modified seed

Step 2: Scanning

Fig. 12 The schematic of the pre-modification method.

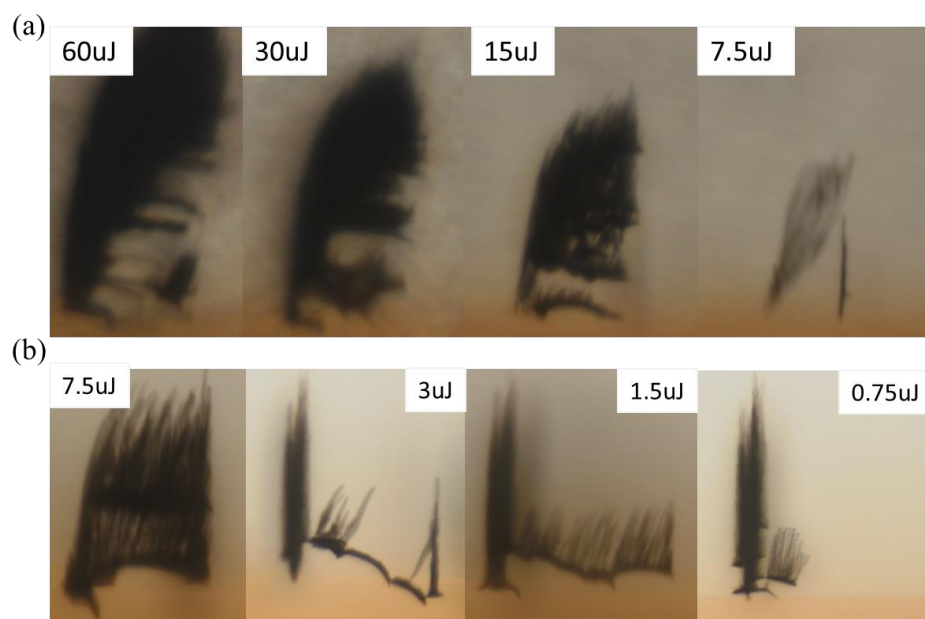


Fig. 13 The effect of the scanning energy to the scanning lines for femtosecond laser scanning (a) without and (b) with pre-modified seed while the scanning speed is fixed to 50 $\mu\text{m/s}$.

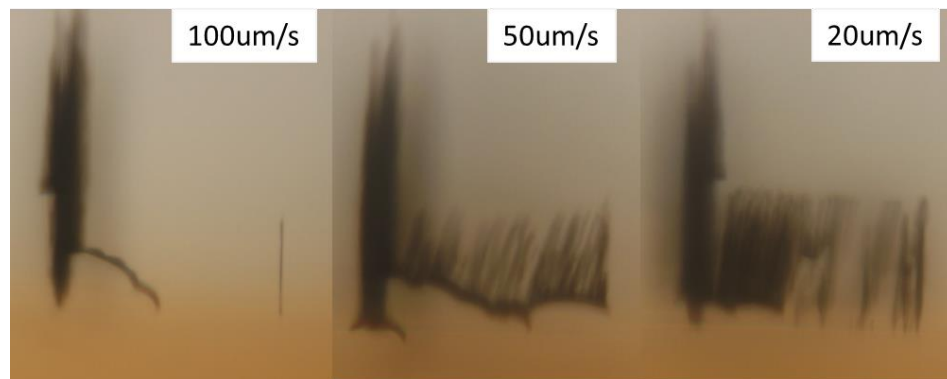


Fig. 14 The effect of the scanning speed to the scanning lines for femtosecond laser scanning with pre-modified seed while the focus energy is fixed to 7.5 μJ and scanning energy is fixed to 1.5 μJ .

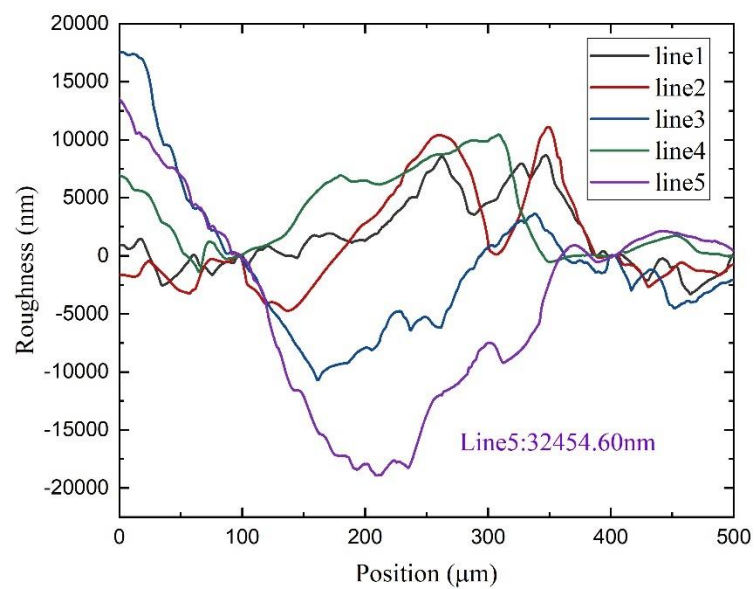


Fig. 15 The roughness of the separated sample by pre-modification method, measured by the Stylus Profilometer.

4. Conclusion

We demonstrate that a single crystal AlN layer grown by Hydride Vapor Phase Epitaxy can be successfully sliced off from an AlN substrate generated by Physical Vapor Transport by scanning a femtosecond laser at the interface. The kerf-loss for the femtosecond laser slicing is significantly smaller than that of conventional slicing method. The separated PVT-AlN substrate can also be reused to grow a new highly pure single crystal AlN with few defects by HVPE after polishing. The multiple spots femtosecond laser slicing and pre-modification was also investigated to improve the efficiency and precision.

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Chapter 3 Femtosecond laser induced multilayer nanograting inside glass

1. Introduction

Femtosecond (10^{-15} s) laser-matter interaction is always an attractive field since the femtosecond laser system was invented due to the extreme high-power density and ultra-short pulse duration. In particular, femtosecond laser micromachining in transparent materials has attracted enormous attention because of the fact that the nonlinear absorption happens when a femtosecond laser is irradiated into the transparent materials, promoting the valence electrons to the conduction band [1-3]. Glass [4-6], crystal [7,8], polymer [9] and other transparent materials have been studied by irradiating femtosecond laser into them. In the nonlinear absorption, with the enough high energy added to the materials, two main excitation processes exist, known as multiphoton ionization and avalanche ionization [10], resulting to the permanent damage. Thanks to these properties, enormous new physical phenomena can be observed, including refractive index change [11], artificial birefringence [12], nanovoid arrays [13], periodic nanostructures [4], and so on.

Among them, one of the most promising phenomena is the femtosecond laser induced self-assembly periodic structures in transparent materials, which named as nanograting, with a width of about 20 nm and a tunable period of 140-320 nm in silica glass [4]. Typically, this femtosecond laser induced nanograting exhibits birefringence. The refractive index difference between extraordinary and ordinary light is about $2-4 \times 10^{-3}$ with a typical period of several ten nanometers which mentioned in Chapter 1 [14]. Since this self-assembly behavior was demonstrated, the ability of nanograting to control the polarization state of transmitted light has been used in various applications, ranging from the polarization-sensitive

waveguides [15,16], polarization converters [17] to optical memories [18,19] and holography [20].

The physical mechanism of the nanograting formation induced by femtosecond laser irradiation is still not completely understood. However, many researchers have tried hard to explain it from various aspects. Some physical models have been proposed in the last ten years [4,21-23]. In the interference model, the nanograting formation is due to the interference of incident laser field with the electron plasma waves [4]. In this model, the electron plasma is generated from the nonlinear ionization induced by the multi-photon absorption, and then the electron plasma concentration becomes periodic in the direction of laser polarization due to the interference between the incident laser and the electron plasma waves, resulting the structural changes in glass. This model can explain many phenomena including the periodicity of the structure and laser energy effect on the period. However, in this model, the high-density plasma waves and high local temperature are necessary, which does not agree well with the later experiments that the nanograting can also generated without high local temperature [24]. Another model [24-27], called transient nanoplasmonics model, shows that when the femtosecond laser is irradiated into the transparent materials, by the interaction between the pulses and the randomly distributed microscopic defects in the materials, localized inhomogeneous nonlinear multi-photon absorption occurs, creating ionization hot-spots. These hot-spots will interact again with the subsequent pulses and transform into spherical nanoplasmas due to the feedback mechanism based on the memory effect of the previous nonlinear ionization [27]. Under the continuous irradiation of the femtosecond laser pulses, these nanoplasmas will gradually grow, and the field enhancement at the edges causes the asymmetric growth of the nanoplasmas along the direction perpendicular to the laser polarization, finally leading to the periodic modulation of the electron plasma concentration. Unfortunately, the lifetime of the

induced nanoplasmas in the focal volume is about 150 fs [28, 29], much shorter than the interval time of adjacent pulses. In addition, many other models are also trying to explain the formation of nanograting with the development of the researches on nanograting. However, as the nanograting can also be found in other complicated materials other than silica glass, conflicts always exist in these models. Thus, further studies are still required to classify the new phenomena.

In this section, to further understanding the formation of nanograting, the multi-layer nanogratings are revealed. The multi-layer nanogratings in our experiments are partially overlapped. The properties of the multi-layer nanogratings are strongly influenced by the polarization direction of the femtosecond laser for different layer. Besides, the birefringence behavior of the nanograting has also an obvious change when controlling the distance between nanograting layers. Specially, in the case of the depth of the second nanograting is deeper than the initial one ($Z > 0 \mu\text{m}$), when the distance between the nanograting layers is $5 \mu\text{m}$, the two-layer nanogratings perform as polarizers. With the distance between the two-layer nanogratings less than $5 \mu\text{m}$, the nanograting formed later will overwrite the first nanograting, while with the distance between the two-layer nanogratings larger than $5 \mu\text{m}$, the two nanograting layers will gradually show their birefringence property independently. The 3 and 4 layers nanograting are also demonstrated to show the special property when the distance between the nanograting layers is $5 \mu\text{m}$. When $Z < 0 \mu\text{m}$, this special distance is $15 \mu\text{m}$.

2. Experiments

The samples we used are synthetic quartz glasses (VIOSIL-SX, Shin-Etsu). The femtosecond laser is a regenerative amplified mode-locked Ti:sapphire laser centered at 800 nm. The pulse duration and the repetition rate are 120 fs and 1 kHz, respectively. The laser pulses are focused into the glass by a $50\times$ objective (Nikon, NA=0.8). A spatial light modulator (LCOS-SLM, X10468-02, Hamamatsu Photonics K.K.) is used to compensate the spherical aberration and modulate the laser into multiple spots or others. The SLM, sample stage, shutter, and laser pulse irradiation are controlled by sending commands from the serial port of a personal computer. After the femtosecond laser is scanned inside the sample, optical polarized microscope is used to measure the birefringence of the structure. The details of the structures are measured by SEM after polishing and etching.

During the multi-layer nanograting formation, the energy of the femtosecond pulses, the pulse number per micrometer, the polarization state of the femtosecond laser and the distance between the layers are controlled to study the birefringence property of multi-layer nanogratings. There are two directions to generate multi-layer nanogratings, top-down ($Z > 0\ \mu\text{m}$) and bottom-up ($Z < 0\ \mu\text{m}$), shown in Fig. 1. Top-down means that after the generation of the first nanograting, the femtosecond laser irradiated through the first nanograting and generate the second nanograting below the first one. On the other hand, bottom-up means that the second nanograting is above the first one. Since the shape of the nanograting from the side view is not a perfect ellipse, the overlap regions of top-down and bottom-up are different, resulting in different phenomena.

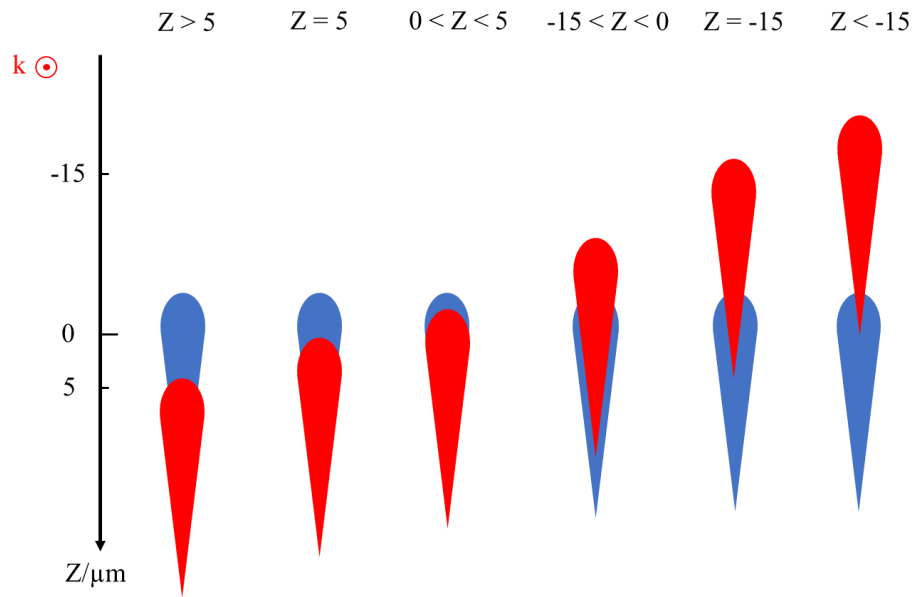


Fig. 1 The schematic of the multilayer nanograting fabrication. The blue shape represents the first nanograting and the red one represents the second nanograting.

3. Results and Discussions

3.1 Single layer nanograting

First, as references, we reproduced the single layer nanograting and overwritten nanograting under different experimental parameters, including the pulse energy, pulse density (scanning speed) and the polarization state of femtosecond laser.

Fig. 2 shows the effect of the pulse energy and the scanning speed to the nanograting formation while the polarization state of the femtosecond laser is perpendicular to the scanning direction. The pulse duration and repetition rate of the femtosecond laser are fixed to 120 fs and 1000 Hz, respectively. The retardance of the nanograting is strongly related to the pulse energy and pulse density. In the later experiments, the pulse energy and scanning speed are chosen to be 1 μ J and 50 μ m/s.

Then the single layer nanogratings are generated with different polarization state of femtosecond laser as shown in Fig. 3 (a). When the polarization state of the femtosecond laser is parallel to the scanning direction, the color of the scanning lines on the polarized microscope is blue and the orientation of the nanovoids of nanograting is perpendicular to the femtosecond laser polarization direction. When the angle between the polarization direction of femtosecond laser and laser scanning direction varies from 0° to 90° , the orientation of the nanovoids is still keep perpendicular to the polarization direction while the retardance is almost the same. The side view of the nanograting when the polarization state of femtosecond laser is parallel to the scanning direction is shown in Fig. 3 (b) (c). A very small red region can be observed at the cross-section of nanograting, which means the periodicity of the structure is very weak. The SEM image can also tell us that the nanostructure at the cross-section of nanograting shows little periodicity.

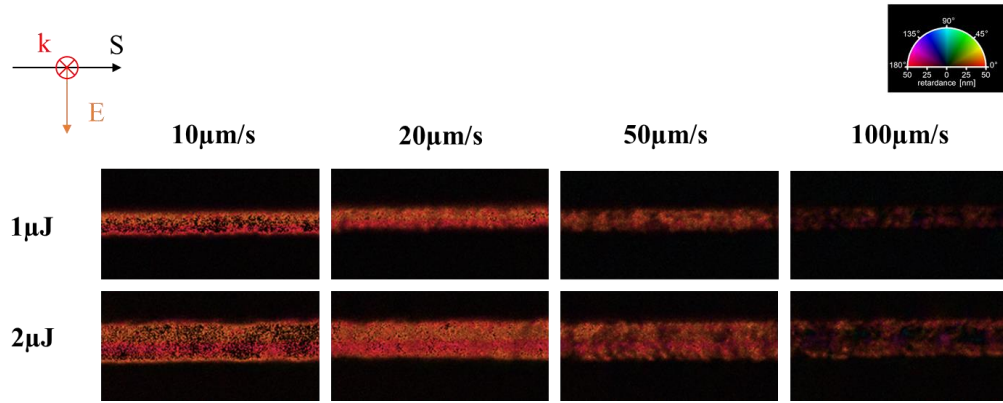


Fig. 2 The effect of the pulse energy and scanning speed to the nanograting formation while the polarization state of the femtosecond laser is perpendicular to the scanning direction measured by polarized microscope.

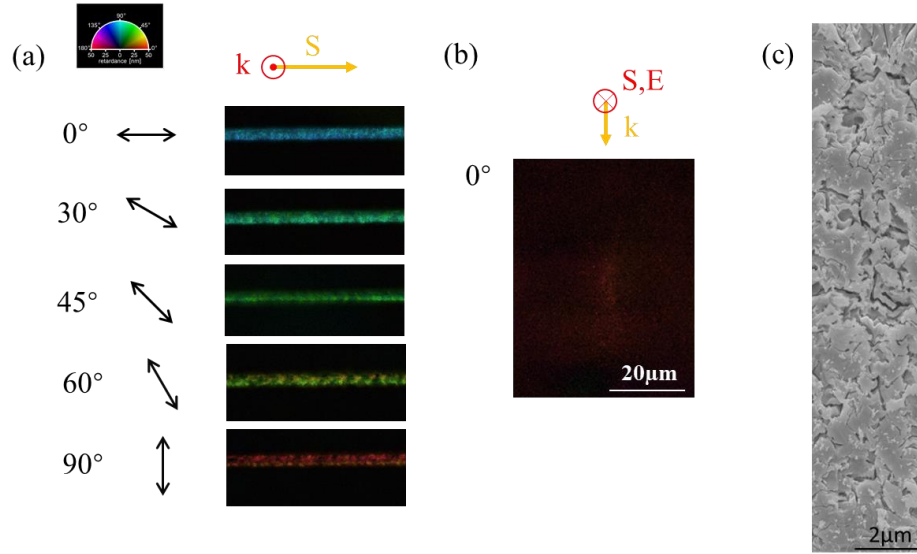


Fig. 3 (a) The effect of the polarization state of the femtosecond laser to the nanograting formation measured by polarized light microscope. The degrees mean the angles between femtosecond laser polarization state and the scanning direction. The color scale indicates the slow axis and retardance of the birefringence. (b) The cross-section of the nanograting while the polarization state of the femtosecond laser is parallel to the scanning direction. (c) The SEM image of (b).

3.2 Overwritten nanograting

Then we try to study the multi-layer nanograting. As a special case, the overwritten nanograting is demonstrated while the distance between the multi-layer nanograting is 0. Previous research has found that the nanograting structure has a rewritable property [24,30]. When the nanograting is rewritten with different scanning direction (perpendicular to the original scanning direction), the previous nanograting can be erased and new nanograting oriented perpendicular to the initial one can be generated. In our experiment, we use femtosecond laser with different polarization direction to scan inside the sample along the same scanning direction. The overwritten nanogratings are shown in Fig. 4 (a), measured by polarized microscope. The initial nanograting is generated by the femtosecond laser whose polarization state is parallel to the scanning direction. Then the nanograting is rewritten at the same position by the femtosecond laser whose polarization state is rotated from parallel to the scanning direction to perpendicular to the scanning direction. The later nanograting almost completely overwrites the initial one. Thus, the birefringence of the structure is determined by the later nanograting.

Fig. 4 (b) shows the cross-section of the overwritten nanograting when the later nanograting is generated by the femtosecond laser whose polarization state is perpendicular to the scanning direction. Unlike to the case, in which the polarization state of femtosecond laser is parallel to the scanning direction, the cross-section of nanograting shows obvious birefringence and is separated into two parts. From the SEM image of the cross-section of nanograting, as shown in Fig. 4 (c), we can know the structure of the cross-section of nanograting. The top of the structure has an obvious and wide periodic area, and the orientation of the nanovoids is perpendicular to the polarization state of femtosecond laser. The middle of the structure shows no birefringence, where the energy density is the highest and can't generate nanograting.

The bottom of the structure also shows the same birefringence as the top structure. However, the region of the periodic structure is much narrower than the top one. The depth of the laser damage region is about 20 μm under the pulse energy as 1 μJ and scanning speed as 50 $\mu\text{m/s}$.

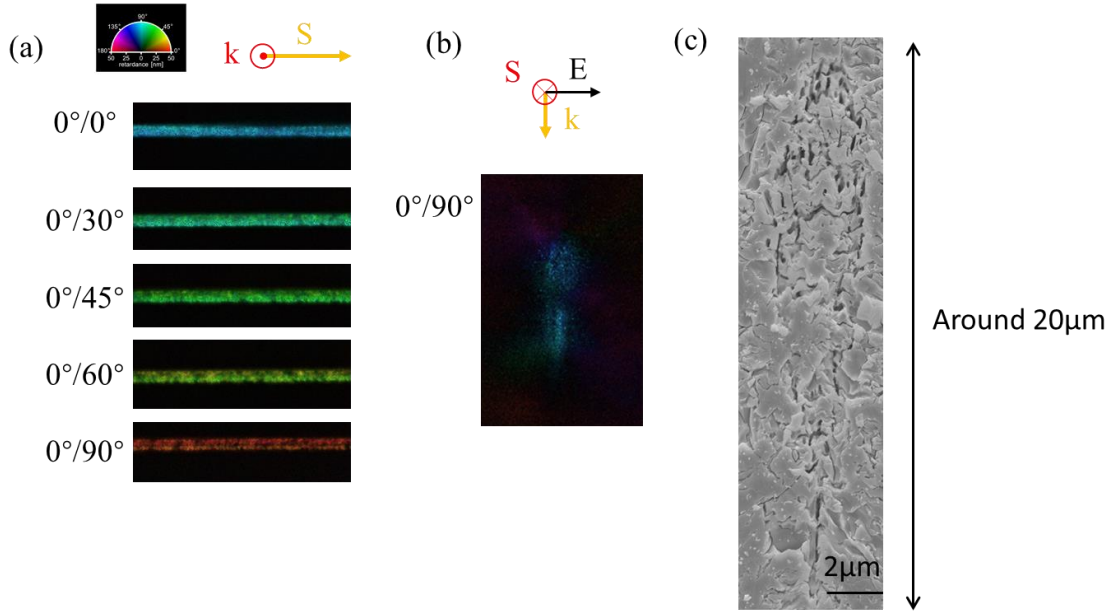


Fig. 4 (a) The overwritten nanograting, while the first nanograting is fixed to 0° and another nanograting is overwritten at the same position with the polarization state from 0° to 90° . (b) The cross-section of the nanograting while the polarization state of the femtosecond laser for overwriting is perpendicular to the scanning direction. (c) The SEM image of (b). The depth of the laser damage region is about 20 μm .

3.3 Double-layers nanograting

The double-layers nanogratings are generated by two directions, top-down ($Z > 0 \text{ } \mu\text{m}$) and bottom-up ($Z < 0 \text{ } \mu\text{m}$). First, we change the distance between the two layers of nanograting by the direction of top-down. The polarization states of the nanogratings here are perpendicular. The results are shown in Fig. 5. Specially, when $Z = 5 \text{ } \mu\text{m}$, the birefringence becomes very weak, almost lower to 0. However, when $Z < 5 \text{ } \mu\text{m}$, the later nanograting just like overwrites the first one. When $Z > 5 \text{ } \mu\text{m}$, the interaction between the nanogratings becomes weak and the nanogratings show their birefringence independently.

The most interesting case is when $Z = 5 \text{ } \mu\text{m}$. Thus, we change the polarization state of the nanogratings when $Z = 5 \text{ } \mu\text{m}$, as shown in Fig.6. The birefringence of the two nanogratings is almost the same and is mainly determined by the later nanograting. This behavior of azimuth is quite similar to the overwritten nanograting. However, the retardance of double-layer nanograting, as shown in Fig. 7, is completely different to the overwritten nanograting, in which the two nanogratings work as polarizers. The top view and cross-section of the double-layer nanograting when the orientations of the nanogratings are perpendicular is shown in Fig. 8. The second nanograting does not overwrite the first one, and the vertical and horizontal nanovoids both exist at the same plane, resulting in the disappearance of anisotropy. However, the specific mechanism of the coexistence of vertical and horizontal nanovoids when $Z = 5 \text{ } \mu\text{m}$ is still not completely understood. To confirm the property of the double-layer nanogratings as polarizers, we also demonstrate the other orthogonal double-layer nanogratings as shown in Fig. 9.

For the $Z < 0 \text{ } \mu\text{m}$, the properties of the double-layer nanogratings are quite different from the ones when $Z > 0 \text{ } \mu\text{m}$. Fig. 10 shows the birefringence of the double-layer nanogratings with different distances between the nanogratings when

the orientations of the nanogratings are perpendicular. The special phenomena occur when the distance between the nanogratings is $15\text{ }\mu\text{m}$, where the birefringence of the lower nanograting becomes weak. Here when $Z > -15\text{ }\mu\text{m}$, the second nanograting which is on the top overwrites the initial one. When $Z < -15\text{ }\mu\text{m}$, the nanogratings seem to be independent. The difference between $Z > 0\text{ }\mu\text{m}$ and $Z < 0\text{ }\mu\text{m}$ is the overlap region of the two nanogratings. When $Z > 0\text{ }\mu\text{m}$, the bottom structure of the initial nanograting is overwritten by the second nanograting. When $Z < 0\text{ }\mu\text{m}$, the top structure of the initial nanograting is overwritten by the second nanograting. Comparing the results of the two methods, we can find that the top structure of the nanograting play an important role on the function of the whole nanograting.

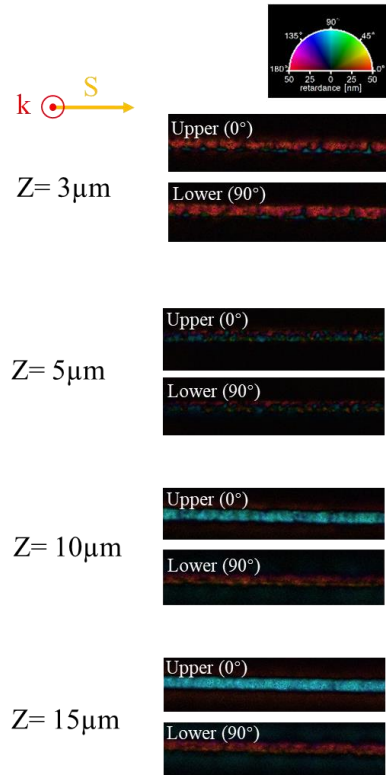


Fig. 5 The double-layer nanogratings with depth while the polarization states for the two layers are perpendicular generated when $Z > 0\ \mu\text{m}$. The upper image shows the nanograting on the top and the lower image shows the nanograting on the bottom.

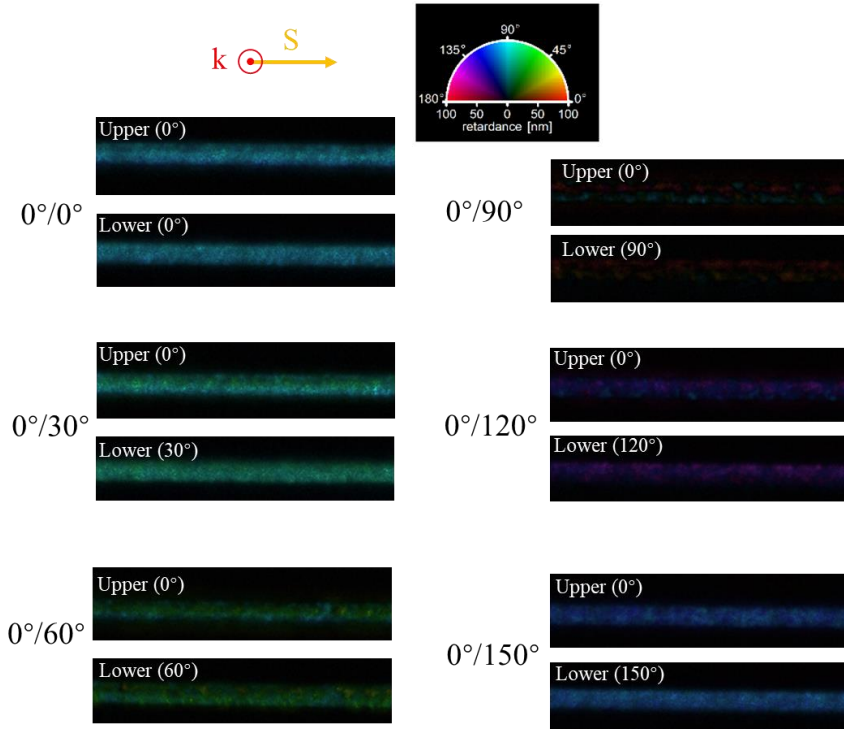


Fig. 6 The double-layer nanogratings generated by the different femtosecond laser polarization state with $Z > 0 \text{ } \mu\text{m}$.

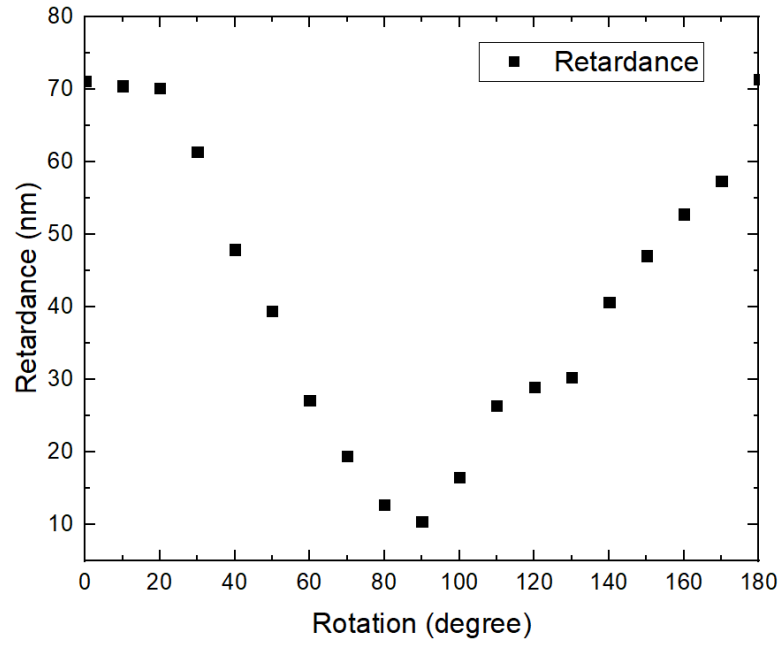


Fig. 7 The retardance of double-layer nanogratings with different femtosecond laser polarization state when $Z > 0 \mu\text{m}$.

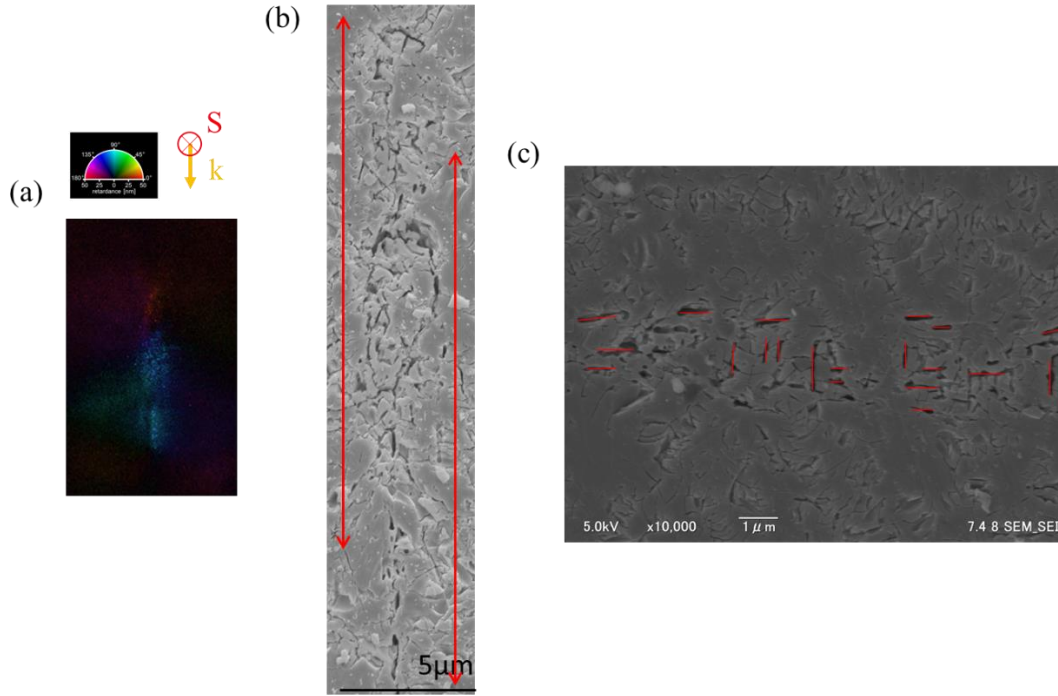


Fig. 8 (a) The cross-section of the double-layer nanogratings when the orientation of the nanogratings are perpendicular with $Z > 0 \mu\text{m}$. (b) The SEM image of (a). The red lines show the lengths of the two nanogratings. (c) The SEM of the top view of the double-layer nanogratings.

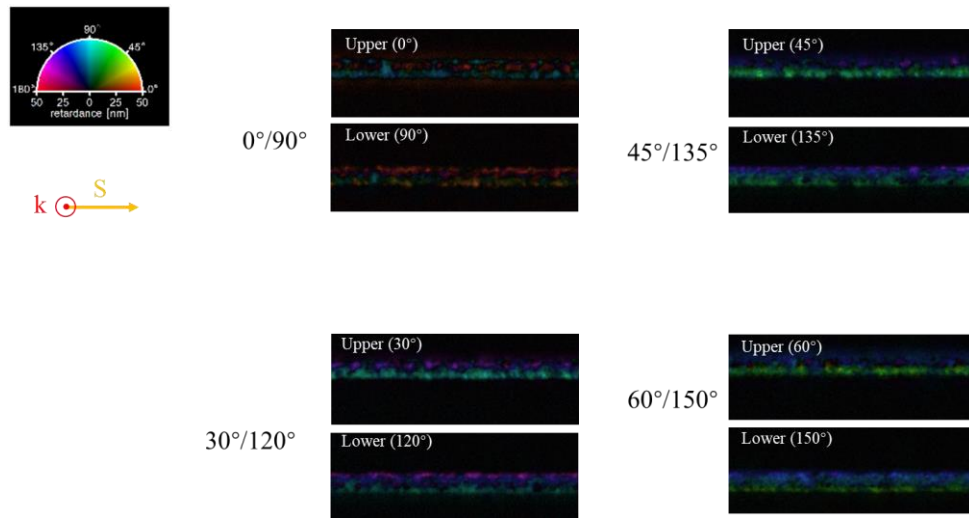


Fig. 9 The double-layer nanogratings generated by different orthogonal femtosecond laser polarization states with $Z > 0 \mu\text{m}$.

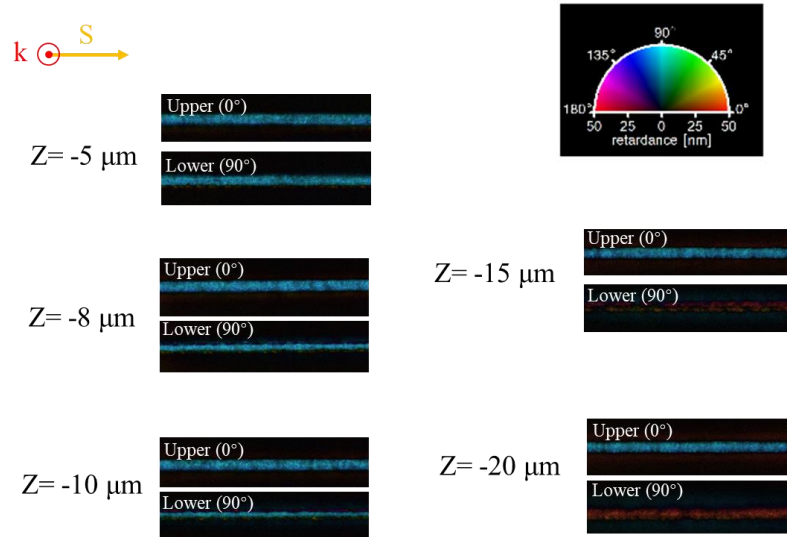


Fig. 10 The double-layer nanogratings with depth while the polarization states for the two layers are perpendicular when $Z < 0 \mu\text{m}$.

3.4 Multi-layers nanograting

The three and four layers nanogratings are also demonstrated. Here we focus on the properties of the multi-layer nanogratings when the polarization states of the irradiated femtosecond laser for different layers are orthogonal. Fig. 11 shows polarized microscope images of the 3-layers nanograting. The 3-layers nanograting generated by top-down direction can't work as polarizers as shown in Fig. 11(a). From Fig. 12(a), we can find that it seems that the third nanograting determine the property of the whole three nanogratings while the first and second nanogratings are generated by the orthogonal polarized femtosecond laser pulse. On the other hand, in the case of bottom-up direction, as shown in Fig. 12(b), the birefringence of the three nanogratings can be observed, although it becomes weak.

The polarized microscope images of the 4-layers nanogratings when the polarization states of the irradiated femtosecond laser for different layers are orthogonal are shown in Fig. 13. The 4-layers nanograting generated by top-down direction shows similar property as the double-layer nanograting.

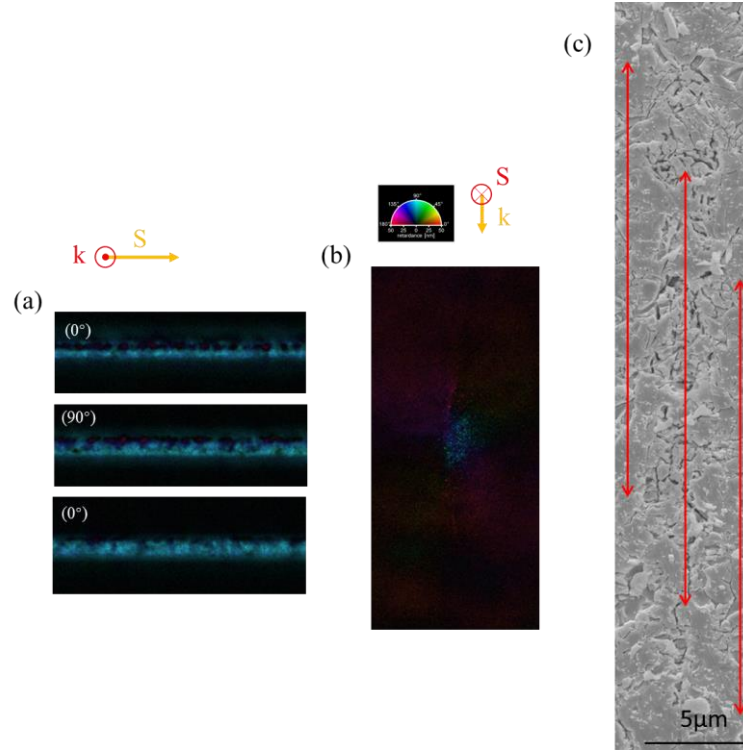


Fig. 11 (a) The 3 layers nanogratings while the polarization states of the irradiated femtosecond lasers are 0° , 90° , 0° from the top to bottom with the fabricated direction of top-down. The distance between the nanogratings is $5\mu\text{m}$. (b) The cross-section of (a). (c) The SEM image of (a). The red lines show the lengths of the two nanogratings.

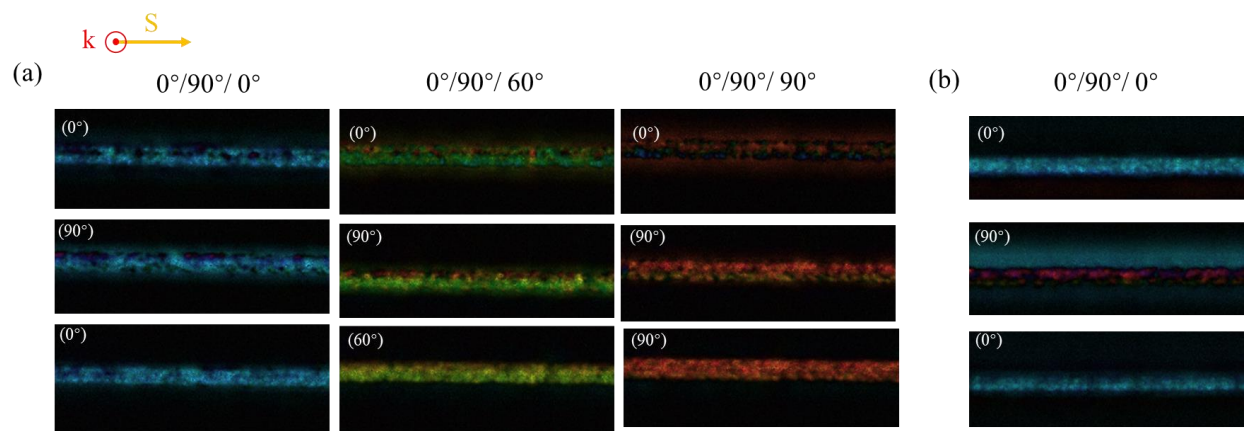


Fig. 12 The 3 layers nanogratings with (a) top-down direction, in which the distance between the nanogratings is $5\ \mu\text{m}$, (b) bottom-up direction, in which the distance between the nanogratings is $15\ \mu\text{m}$.

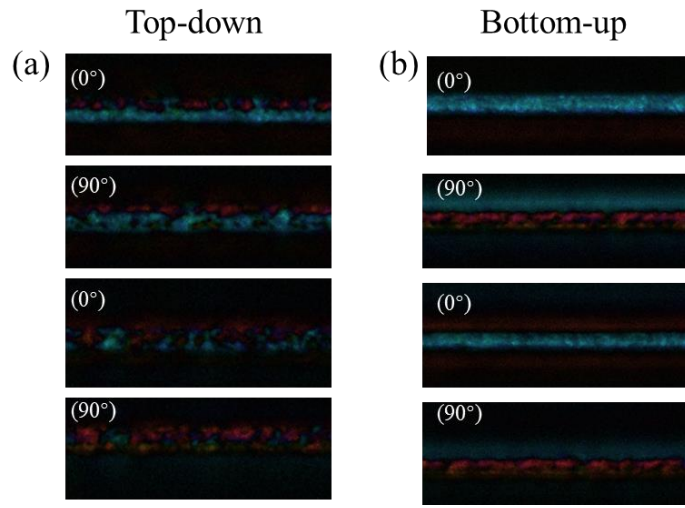


Fig. 13 The 4 layers nanogratings while the polarization states of the irradiated femtosecond lasers are 0° , 90° , 0° , 90° from the top to bottom with (a) top-down, in which the distance between the nanogratings is 5 μm , (b) bottom-up, in which the distance between the nanogratings is 15 μm .

4. Conclusion

We demonstrate the multi-layer nanogratings to further understanding the formation of nanogratings. Pulse energy, pulse density, polarization of the incident laser and the distance between the multi-layer nanogratings are controlled to explore the formation of multi-layer nanogratings. We generate the multi-layer nanograting with two directions, top-down and bottom-up, due to the structure difference of the nanograting along the laser irradiation direction. The birefringence of the multi-layer nanograting directly related to the polarization state of the incident laser. Specially, in the top-down direction, when the distance between multi-layer nanogratings is 5 μm , the 2 and 4-layers nanogratings work as polarizers and when the distance between multi-layer nanogratings is less than 5 μm , the later nanograting will overwrite the initial one. The disappearance of the birefringence when the distance between the multi-layer nanogratings is 5 μm is mainly due to the coexistence of vertical and horizontal nanovoids. In the bottom-up direction, this special distance between multi-layer nanograting is 15 μm , where the birefringence of the initial nanograting becomes weak with the orthogonal polarization states of the incident lasers. The phenomena may mainly due to the structure of the cross-section of the nanograting, whose top structure is quite different to the bottom structure. However, further studies are required to completely understanding the special phenomena when the distance between multi-layer nanogratings is 5 μm .

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Chapter 4 Simulation of metasurface with rectangular pillars and triangular pillars and the effect of the rounded angles on the sharp corners of these pillars

1. Introduction

One of the recent interesting application of femtosecond laser direct writing is the fabrication of metasurface, due to the highly precise modification as small as 20 nm on the surface of material [1].

Metasurfaces, composed of specifically designed subwavelength units (meta-atoms) in a two-dimensional plane, offer a new paradigm to manipulate every aspects of light that show great potentials for miniaturizing optical systems. By adjusting the shapes, sizes, orientations or positions of meta-atoms, the phase gradient is introduced at the interface for arbitrary molding of the optical wavefront [2-5].

Several metasurface platforms have been explored to replicate the functionality of common optical components such as wave retarders, polarizers, beam splitters, lenses, and focusing mirrors. However, none of these platforms achieve complete control over both polarization and phase with subwavelength sampling and high transmission. A metasurface platform achieves complete control over polarization and phase if it can generate any desired physically admissible spatially varying polarization and phase distributions from an input beam with arbitrary polarization and phase distributions. The plasmonic metasurfaces are suffering from limited efficiencies due to fundamental limits [6,7] and metal absorption losses [8-10]. However, the metasurface platform based on high-contrast dielectric meta-atoms can provide complete control of polarization and phase with subwavelength spatial resolution and the efficiency can be as high as 97% [11]. Due to the high refractive

index of the meta-atoms can confine the light inside them, resulting in the independence between the meta-atoms.

One of the most attractive application of metasurface is the metasurface holography. Metasurface holograms, utilizing subwavelength meta-atoms, can achieve a remarkable subwavelength resolution of 350 nm under visible light. In addition, the metasurface holograms can offer an enlarged viewing angle, higher space-bandwidth product (SBP), and suppressed higher-order diffraction in the far field [12]. Compared with currently commercially available wavefront modulation devices and other non-metasurface wavefront modulation devices that are not commercially available, metasurfaces perform very well [13].

To generate a metasurface hologram, the geometric parameters of the meta-atoms on the metasurface should be carefully designed. The ability to completely control the phase, amplitude and polarization of light by metasurfaces is very sensitive to these geometric parameters. However, by current fabrication methods, such as femtosecond laser direct writing, it is difficult to generate the meta-atoms with accurate parameters in nanoscale, especially the sharp corners of the meta-atoms. These deformations may strongly influence the functions of metasurface. To make the fabrication easier, many studies introduced the fillets (rounded corners) to the corners of meta-atoms or used the elliptical meta-atoms [11,14].

In this section, we numerically demonstrated the dielectric metasurface with triangular pillars and isosceles rectangular pillars. Then we introduced the fillets to the sharp corners of these pillars. In the simulations, by controlling the radius of the fillet to a specific value, we can find the bound state in continuum (BIC) and quasi-BIC, which have great potential for the super absorbers [15], sensing [16,17], optical nonlinearity enhancement [18] and so on. We believe our studies on the fillets of the sharp corners of meta-atoms can provide a guide to generate accurate metasurface

hologram and the BIC caused by controlling the radius of the fillets can offer a novel perspective on generating BIC.

2. Structure Design and Method

To simulate the dielectric metasurface, we use TiO₂ (the refractive index is 2.61) as the material of the meta-atoms and glass (the refractive index is 1.45) as the substrate by the COMSOL MultiPhysics. The model of the structure is shown in Fig. 2. Perfect matched layer (PML) is used to simulate the boundary conditions at the input and output port and the periodic boundary conditions are applied to the other surface. In the inset, T means the height of the pillar; P means the size of the unit cell; W and H means the width and length of the pillar; α means the rotation of the pillar. The high refractive index pillars here can confine the light inside them, reducing the interferences from their neighbouring pillars as shown in Fig. 3. We simulate two types of meta-atoms, rectangular pillars and isosceles triangular pillars. To make sure the high efficiency, for both the rectangular pillar and the isosceles triangular pillar, the height of the pillars and the size of unit cell are fixed to 600 nm and 350 nm at the wavelength of 532 nm, respectively. By carefully designing geometric parameters of the pillars, including the width W, height H and rotation θ as shown in Fig. 4, we can control the phase shifts after the light propagating through the pillars covering the range from 0 to 2π with almost the same amplitude, which is necessary for the metasurface hologram. Generally, eight meta-atoms with the phase shifts in $\pi/4$ increments are required [19].

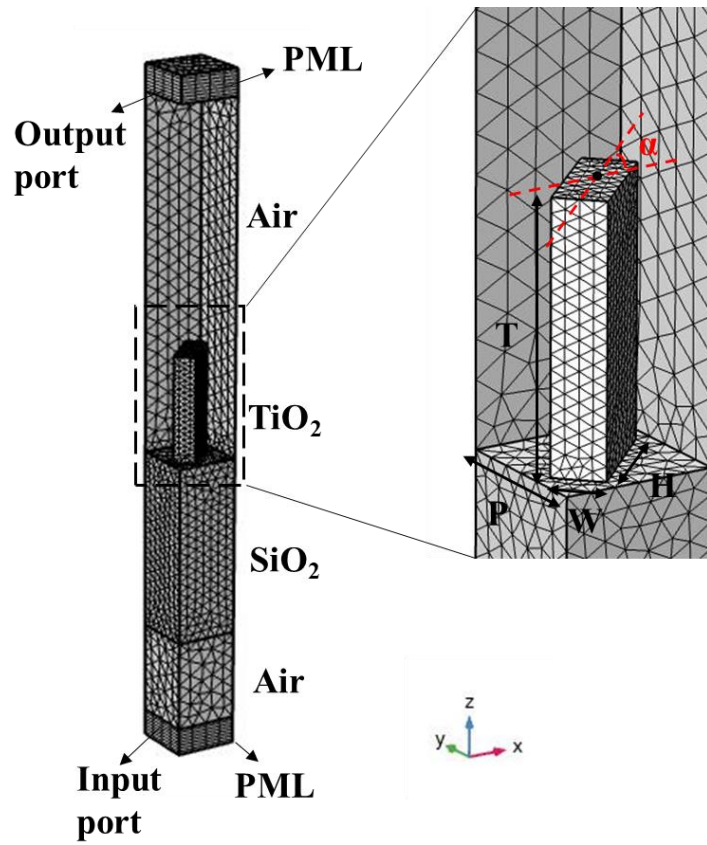


Fig. 2 The model of the meta-atoms.

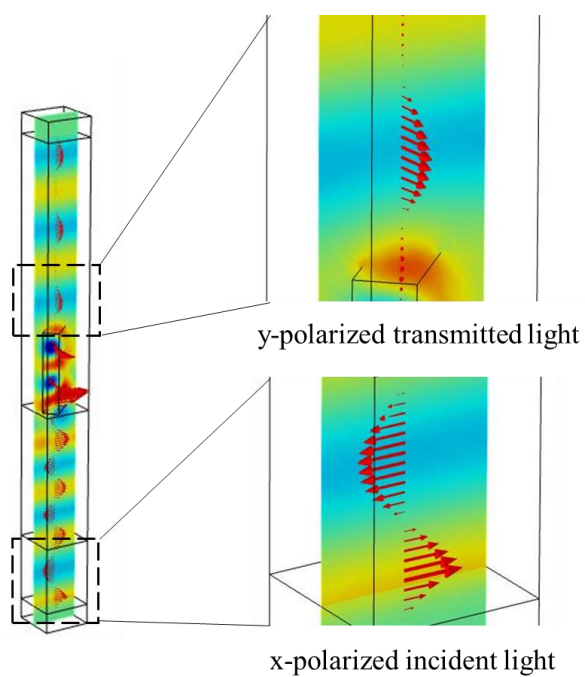


Fig. 3 The simulated transmission of light through meta-atom, which works as a half-wave plate.

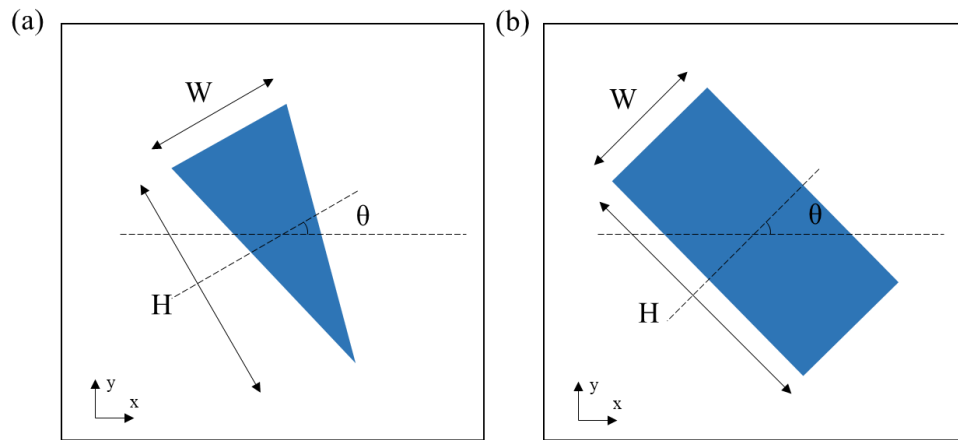


Fig. 4 Design of the meta-atoms. The height of the pillars is 600 nm and the size of the unit cell is 350nm. The pillars are set at the center of the unit cell. (a) The top view of the isosceles triangular pillar determined by the height H , width W and rotation θ . (b) The top view of the rectangular pillar determined by the height H , width W and rotation θ .

3. Results and Discussions

3.1 Simulation for rectangular pillars

The rectangular pillars have been studied a lot [20], that can be designed working as half-wave plates. We simulate the rectangular pillars with different geometric parameters, including the height H , width W and rotation θ . As mentioned before, the rectangular pillars here can work as half-wave plates, thus we can obtain the y-polarized transmitted light by changing θ to 45° . When θ is fixed to 45° , the amplitude and phase shift of the y-polarized transmitted light is shown in Fig. 5. After the overall simulations, 8 pillars are chosen as shown in Table 1 and Fig. 6(a) to cover 2π . The θ of these 8 pillars are fixed to 0° , at which the transmitted light is x-polarized while the incident light is also x-polarized. The half-wave plate behavior of the rectangular pillar can be seen in Fig. 6(b).

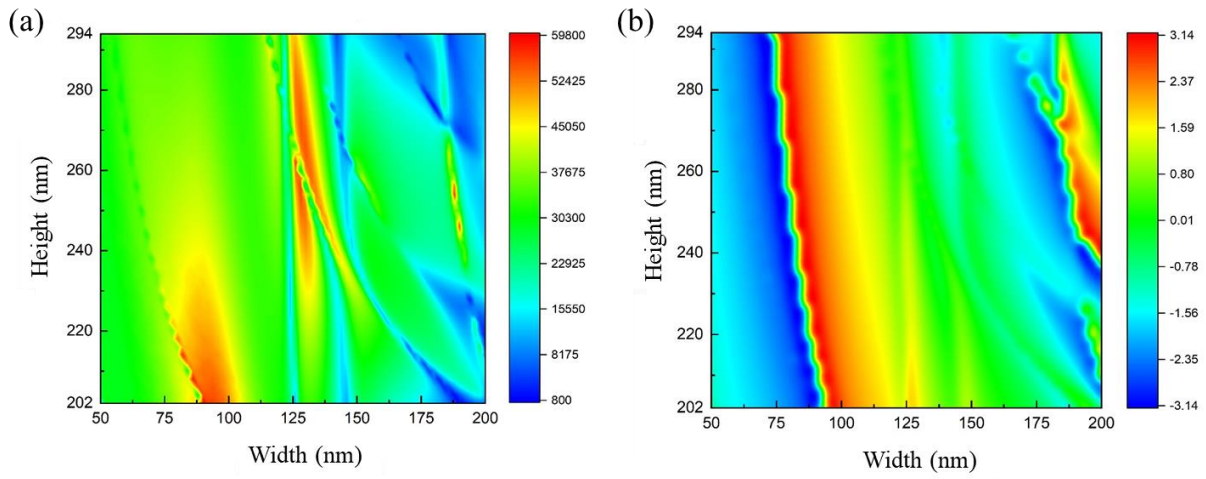


Fig. 5 (a) The amplitudes and (b) phase shifts of the y-polarized transmitted light after propagating through the rectangular pillar as a function of the width and height of the pillar.

Table 1 The geometric parameters of the designed rectangular pillars

Pillar number	1	2	3	4	5	6	7	8
Height/nm	246	214	194	186	112	108	100	86
Width/nm	112	108	100	86	246	214	194	186
Phase shift /rad	-2.81	-2.02	-1.18	-0.38	0.28	1.20	2.14	2.81

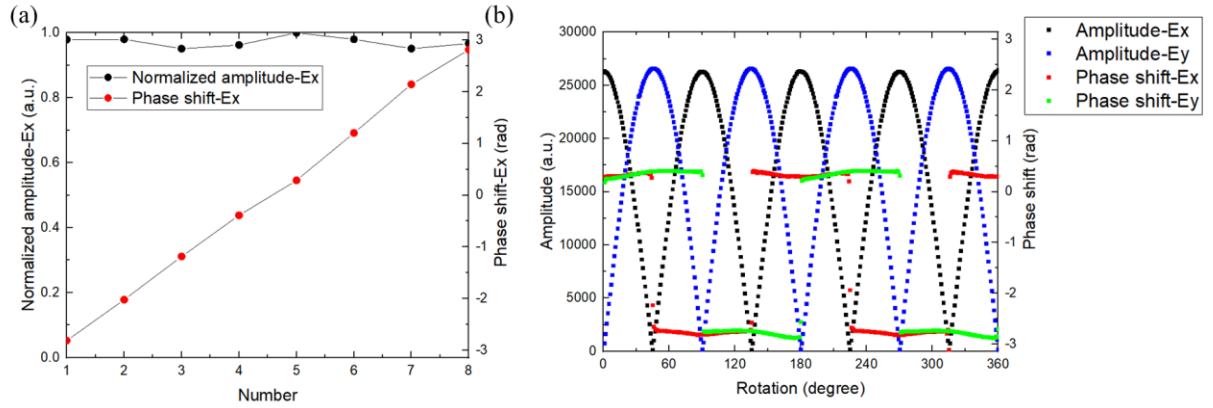


Fig. 6 (a) The amplitudes and phase shifts of the x-polarized transmitted light after propagating through the eight chosen rectangular pillars for the x-polarized incident light. (b) The half-wave plate behavior of the rectangular pillar 1 in Table 1.

3.2 Simulation for isosceles triangular pillars

On the other hand, the isosceles triangular pillars were rarely studied. Here we use the same method as the rectangular pillars to simulate the isosceles triangular pillars. The geometric parameters of isosceles triangular pillars we controlled are height H , width W and rotation θ . We also choose 8 pillars, as shown in Table 2 and Fig. 7(a), covering 2π while the transmitted light and incident light are both x-polarized. Unlike to the rectangular pillars, the isosceles triangular pillars can't be designed as half-wave plates. Fig. 7(b) shows the amplitude and the phase shift of the transmitted light while rotating the isosceles triangular pillar 1 in Table 2. Although the isosceles triangular pillars don't have center symmetry, the modulations to the transmitted lights are almost the same for the rotation of θ and $\theta+180^\circ$. Besides, for the rotation of θ ($<180^\circ$) and $180^\circ-\theta$, the modulations to the x-polarized transmitted lights are also the same and the modulations to the y-polarized transmitted light have a phase shift of π . Therefore, the modulation of the isosceles triangular pillars to the transmitted light is strongly according to the rotation of the axis of symmetry.

Table 2 The geometric parameters of the designed isosceles triangular pillars

Pillar number	1	2	3	4	5	6	7	8
Width/nm	200	300	200	120	280	280	260	300
Height/nm	180	130	100	190	210	170	150	150
Rotation/deg	90	90	0	0	0	0	0	90
Phase shift /rad	-2.35	-1.57	-0.79	0	0.80	1.59	2.36	-3.13

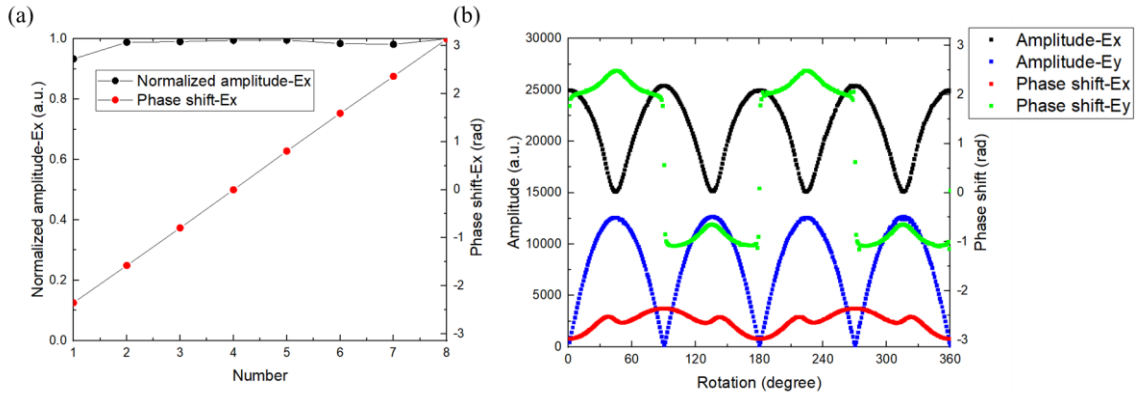


Fig. 7 (a) The amplitudes and phase shifts of the x-polarized transmitted light after propagating through the eight chosen isosceles triangular pillars for the x-polarized incident light. (b) The amplitude and phase shift of the transmitted light for the isosceles triangular pillar 1 in Table 2, as a function of θ .

3.3 The effect of the fillets on the sharp corners

After finding the 8 pillars whose phase shifts cover the whole 2π , we introduce the fillets to the sharp corners of these pillars as shown in Fig. 8. In the case of the rectangular pillars, we introduce the fillets to 1 corner, 2 diagonal corners, 2 mirror symmetric corners, 3 corners, and 4 corners. As for the isosceles triangular pillars, we introduce the fillets to 1 corner, 2 symmetric corners, 2 asymmetric corners, and 3 corners. The symmetric condition of each case is different, resulting in the different effect to the amplitude and phase shift. The sizes of the fillets are changed from 1 nm to the maximum to study the effect of the fillets.

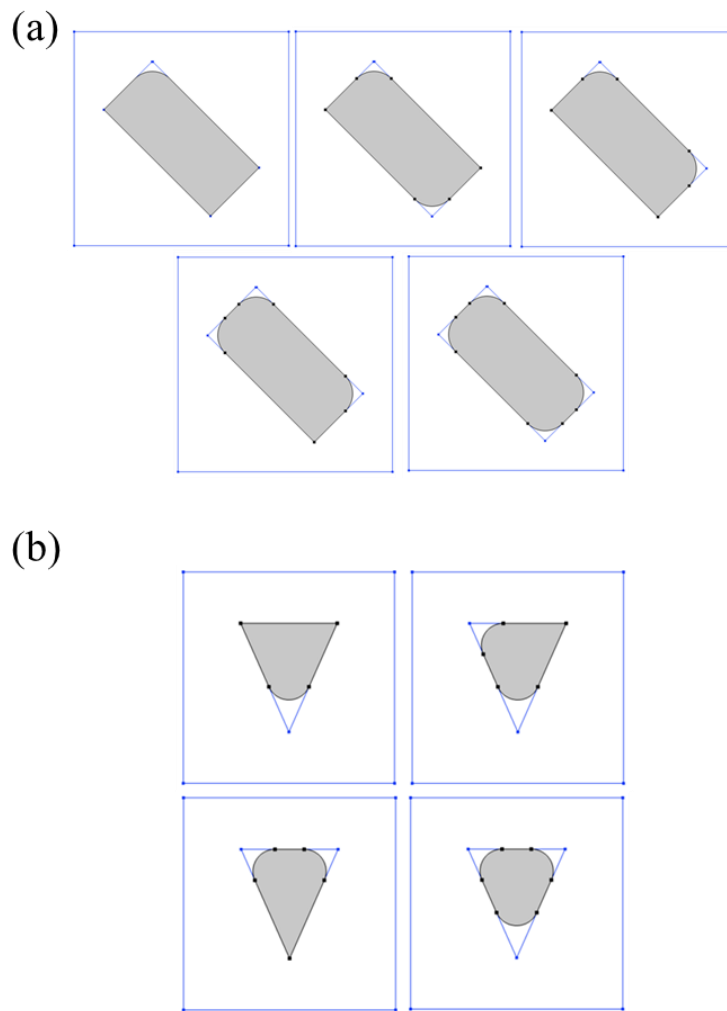
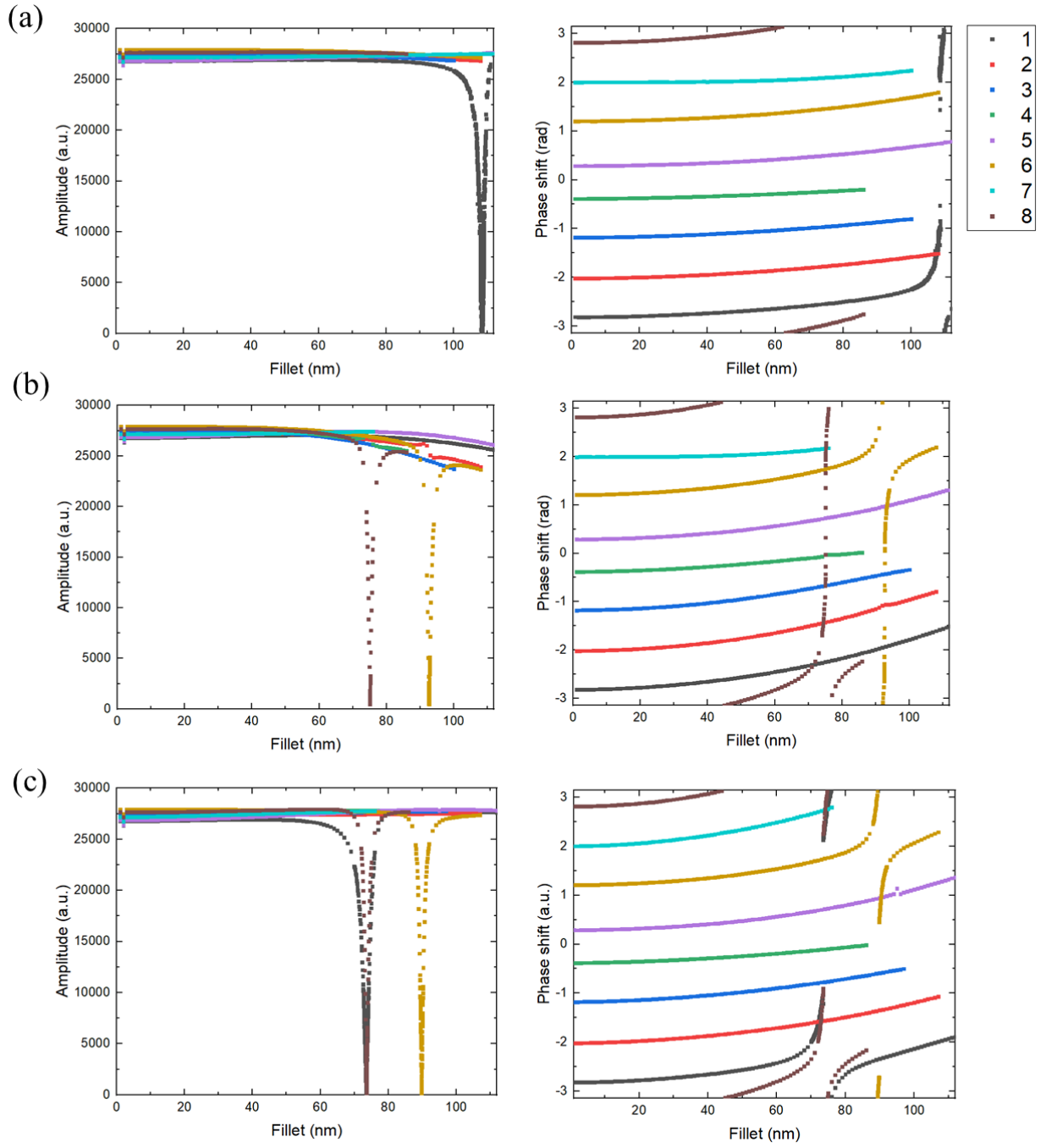


Fig. 8 The fillets we introduce to (a) rectangular pillars and (b) isosceles triangular pillars.

Fig. 9 shows the effect of the fillets to the amplitudes and phase shifts of the transmitted lights under different symmetric conditions for rectangular pillars. With the increase of the size of the fillets, the amplitudes of transmitted lights almost keep the same, while the phase shifts of the transmitted lights are gradually increasing except for some special fillet radii in Fig. 9 (a), (c), (d) and (e), which indicating the 1 fillet, 2 symmetric fillets, 3 fillets and 4 fillets. However, at some special fillet radii in Fig. 9 (a), (b) and (c), the amplitude of the transmitted light drops to 0 and the phase shift has an abrupt change. This is because of the BIC controlled by the parameter tuning [21]. Typical results owing to BIC is shown in Fig.10 when 2 symmetric fillets are introduced to pillar 6. The transmittance when the fillet radius equal to 89.76 nm is almost 0, which means the energy of light is completely confined in the pillar as shown in Fig. 11 (a). Around the special fillet radii of BIC, the quasi-BICs exist, the energy is not completely confined in the pillar, but radiates to the far field. The phase shift of the transmitted light around the BIC has an abrupt change, however the trend of the phase shift is still kept if ignoring the BIC and quasi-BIC. The half-wave plate behavior of the rectangular pillar is also disappeared around the parameters for BIC as shown in Fig. 11 (b).

Specially, in the case of 2 diagonal fillets on the rectangular pillars, the amplitude of the transmitted light begins to reduce when the radius of the fillet is near the maximum. This is due to the deformation of the width and the length of the rectangular pillar, which resulting in the leak from x-polarized light.



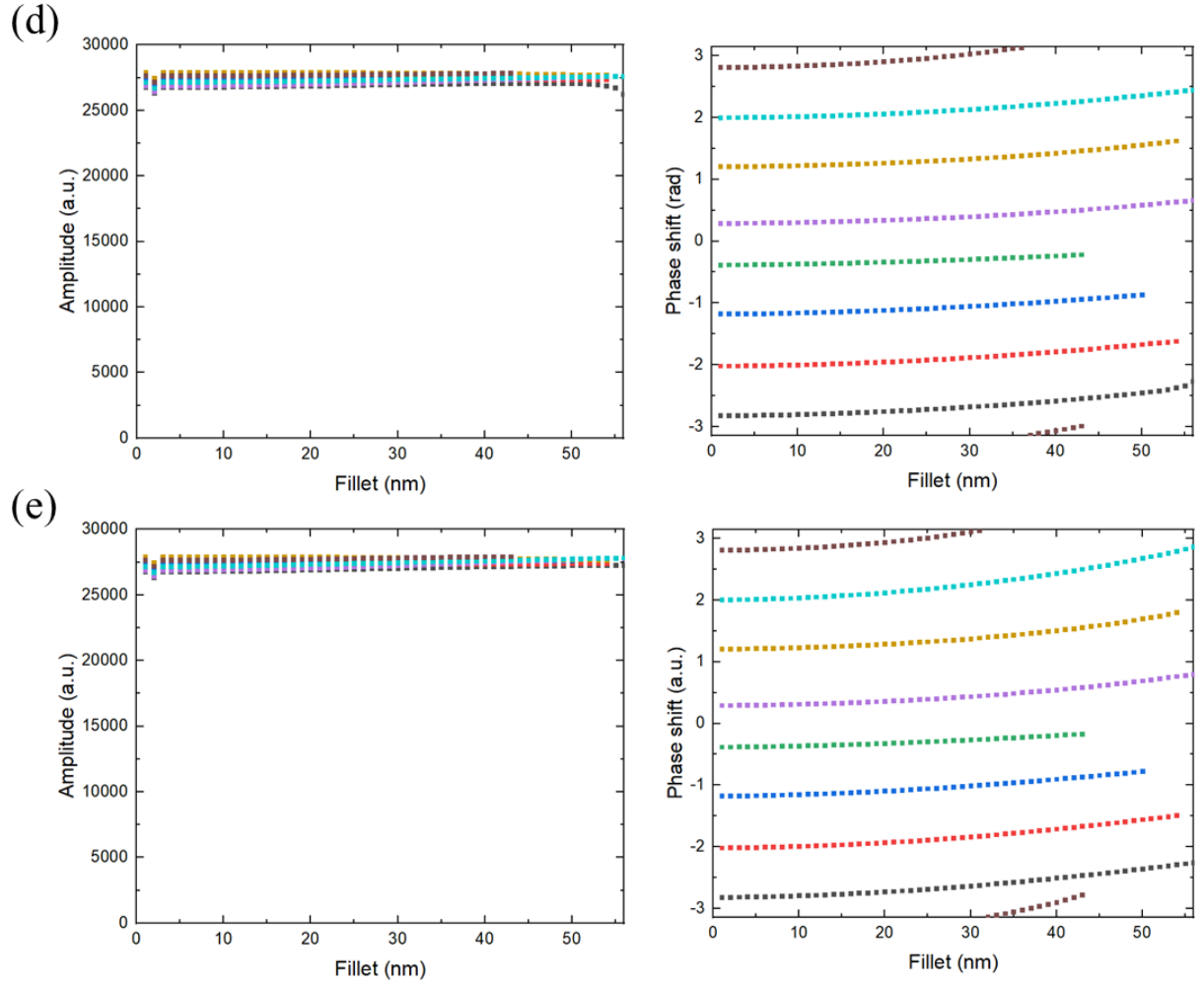


Fig. 9 The effect of the fillets to the amplitudes and phase shifts of the transmitted light for (a) 1 fillet, (b) 2 diagonal fillets, (c) 2 symmetric fillets, (d) 3 fillets and (e) 4 fillets on rectangular pillars.

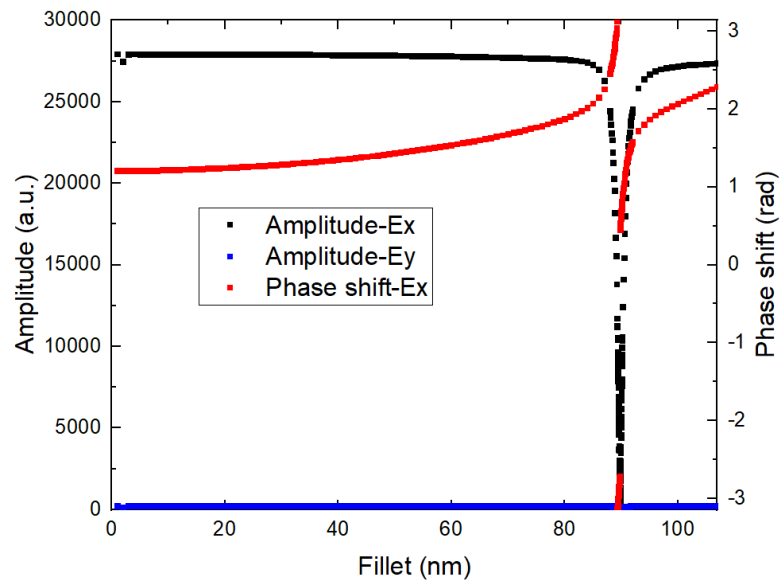


Fig. 10 The amplitude and phase shift variation with the fillet in the case of 2 symmetric fillets on pillar 6.

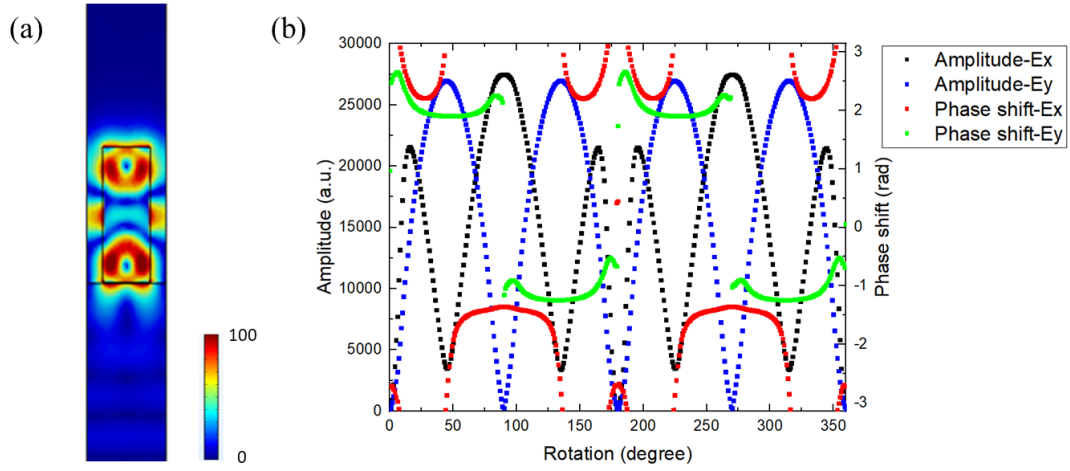
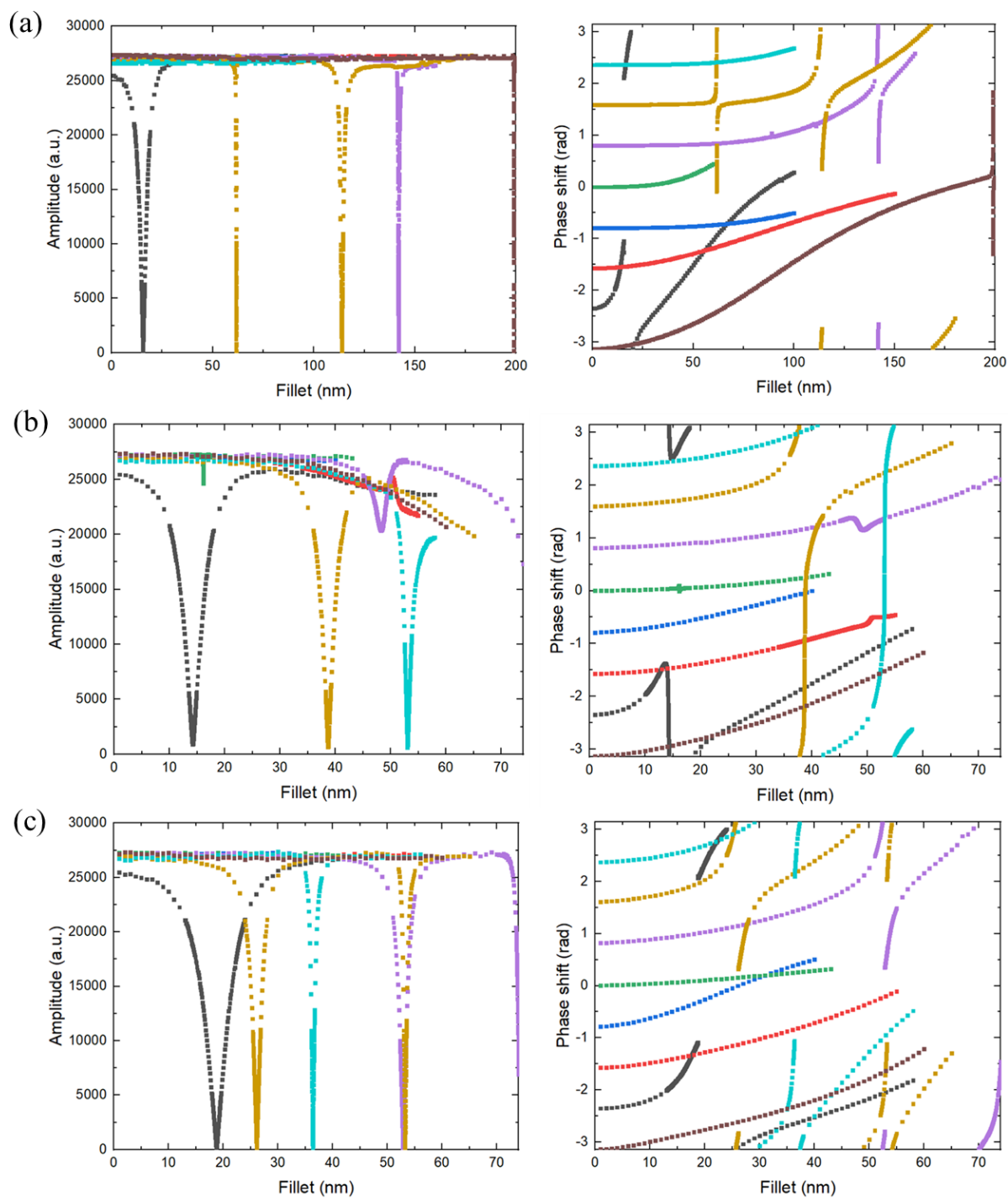


Fig. 11 (a) The energy density when x-polarized light propagates through the rectangular pillar 6 with 2 symmetric fillets from the bottom to the top at the fillet radius of 89.76 nm. (b) The amplitude and phase shift of the transmitted light propagating through the rectangular pillar 6 with 2 symmetric fillets at the fillet radius of 89.76 nm, as a function of θ .

The effect of the fillets to the amplitudes and phase shifts of the transmitted lights for isosceles triangular pillars is shown in Fig. 12. Similar to the case of rectangular pillars, the BIC and quasi-BIC also exist at some special fillet radii. The amplitude almost keeps the same and the phase shift increases gradually with the increase of fillet radius if ignoring the BIC and quasi-BIC in Fig. 12 (a), (c) and (d). Specially, the amplitude of the transmitted light can't keep the same when the radius of the fillet is near to the maximum in Fig.12 (b), in which the fillets are introduced to 2 asymmetric corners of the isosceles triangular pillars. This is mainly due to the break of the axis of symmetry of the isosceles triangular pillar. Comparing to the rectangular pillars, it is much easier to find the BIC in isosceles triangular pillars in our simulation. Besides, even the radius of the fillets is small to about 10 nm, the quasi-BIC still exists, such as the pillar 1 in Fig. 13 (the black spots). That means a small error of the geometric parameters for isosceles triangular pillars may induce a large amplitude and phase change of the transmitted light.



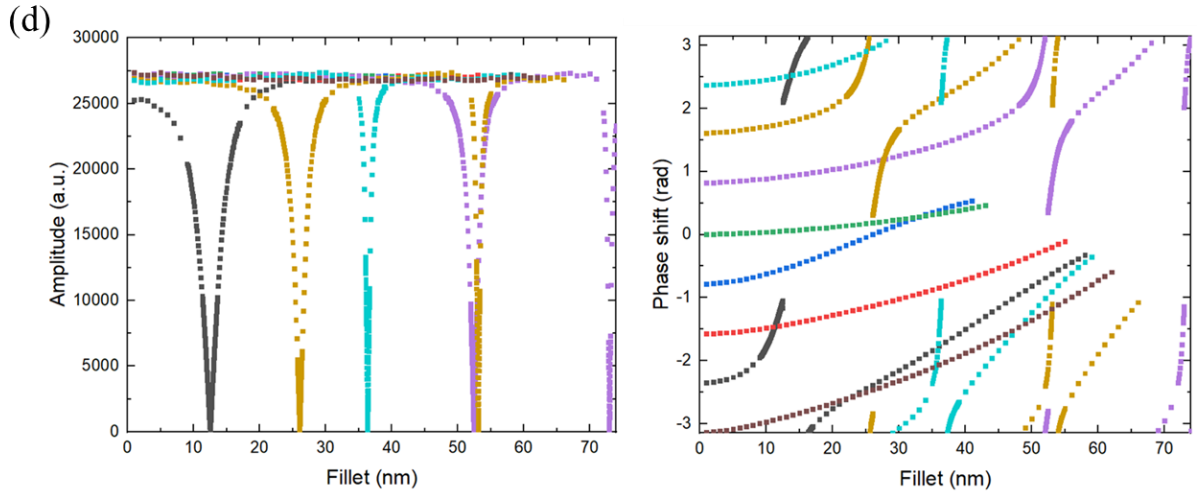


Fig. 12 The effect of the fillets to the amplitudes and phase shifts of the transmitted light for (a) 1 fillet, (b) 2 asymmetric fillets, (c) 2 symmetric fillets and (d) 3 fillets on isosceles triangular pillars.

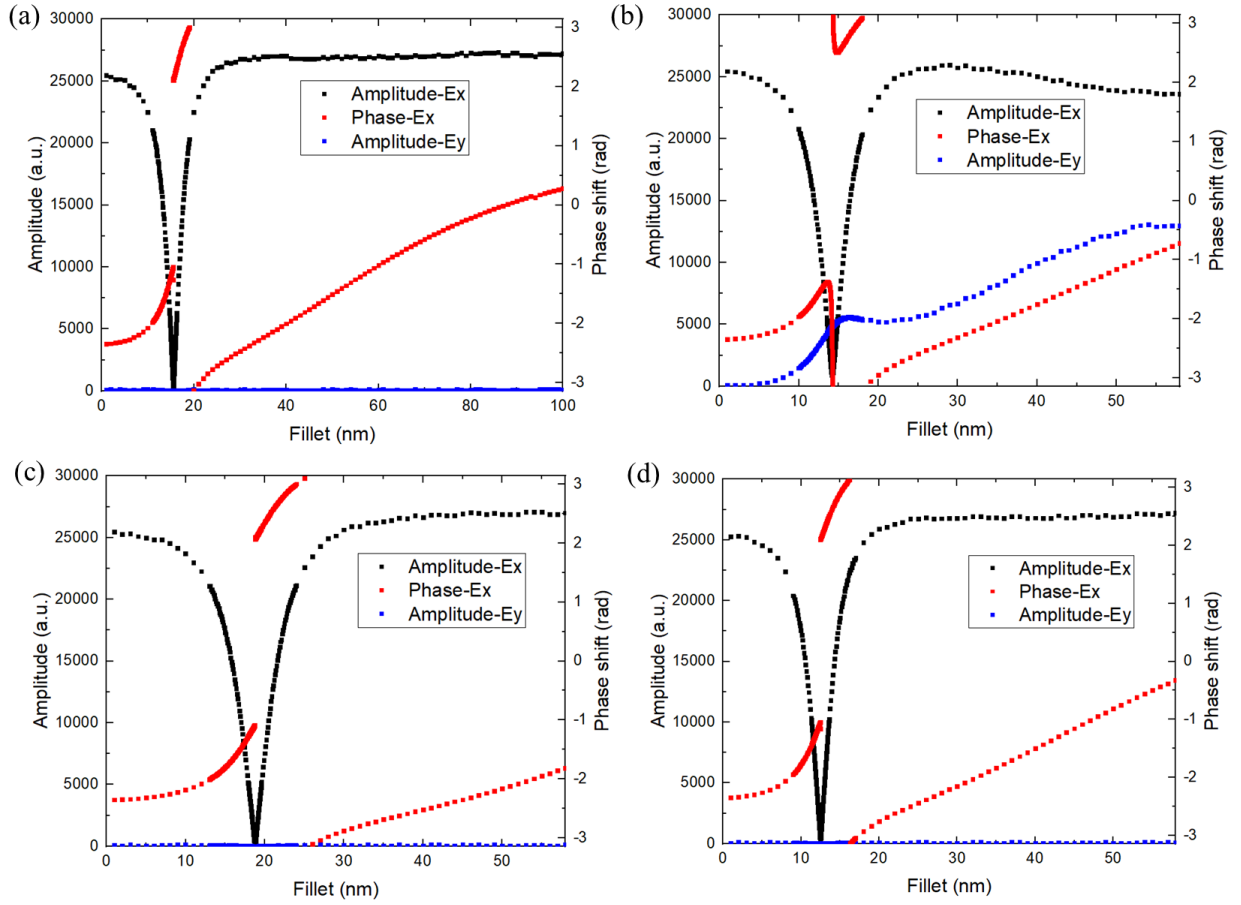


Fig. 13 The amplitude and phase shift variation with the fillet in the case of (a) 1 fillet, (b) 2 asymmetric fillets, (c) 2 mirror symmetric pillar, and (d) 3 fillets on isosceles triangular pillar 1.

4. Conclusion

In conclusion, we numerically study the dielectric metasurface with meta-atoms of rectangular shape and isosceles triangular shape. We choose 8 kinds of shapes for both rectangular pillars and isosceles triangular pillars to cover the whole 2π of the phase shift of the transmitted light. The rectangular pillars can work as half-wave plate, however the isosceles triangular pillars can't. In contrast, the modulation of the isosceles triangular pillar is strongly related to the axis of symmetry of the pillar. We also introduce the fillets to the corners of the rectangular pillars and isosceles triangular pillars with different symmetric condition. For both the rectangular pillars and isosceles triangular pillars, we can find the bound state in continuum (BIC) while changing the radius of the fillets. Except the BIC and quasi-BIC, the amplitudes of the transmitted lights are almost the same and the phase shifts are always increased gradually with the increase of the radius of the fillets. However, for rectangular pillars, the fillets introduced to 2 diagonal corners can make the energy of x-polarization leaking to other states when the radius of the fillets is near the maximum. For triangular pillars, the fillets introduced to 2 asymmetric corners can also reduce the energy of x-polarized light. Both of them are caused by the break of the original symmetry. These meta-atoms with fillets can find potential applications on the generation of accurate metasurface hologram, metasurface absorber, sensing and so on.

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Publication list

Chapter 2

‘Femtosecond Laser Slicing of Single Crystal AlN Layer Grown by Hydride Vapor Phase Epitaxy on an AlN Substrate Generated by Physical Vapor Transport’,

Y. Yang, K. Matsui, Y. Shimotsuma, T. Furuya, R. Yamamoto, T. Nagashima, M. Shimizu, and K. Miura,

Journal of Laser Micro/Nanoengineering, **19**, (2024) 63-66.

Chapter 3

‘The effect of rounded angle on the sharp corners of meta-atoms in dielectric metasurface’,

Y. Yang, Y. Shimotsuma, M. Shimizu, and K. Miura,
(under review)

Chapter 4

‘Femtosecond laser pulses induced multi-layer nanogratings in glass’,

Y. Yang, Y. Shimotsuma, M. Shimizu, and K. Miura,
(to be submitted)

Other publications

‘Influence of experimental parameters on characteristics of focusing light through scattering media by wavefront shaping’,

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Physical Review A, **104**, 5(2021) 053535.

Acknowledgements

The present thesis has been carried out under the direction of Professor Kiyotaka Miura in Graduate School of Engineering in Kyoto University.

First of all, I would like to express my deepest appreciation to Professor Kiyotaka Miura, who supervised this work, for his kind encouragement, insightful guidance, constructive suggestions, and valuable advices throughout the course of this study. I'm also deeply indebted to Professor Katsuhisa Tanaka and Professor Koji Fujita for their valuable comments and discussions.

I would also like to extend my deepest gratitude to Associate Professor Yasuhiko Shimotsuma and Assistant Professor Masahiro Shimizu for their perseverant guidance and constructive advices from the selection of the thesis topic to the realization of the experiments.

I would also like to thank Mr. Taishi Furuya, Mr. Reo Yamamoto, Mr. Toru Nagashima from Tokuyama corporation for their support on the samples and measurements;

I show special thanks to all the members in Miura laboratory for their supports throughout the research study.

I also acknowledge Dr. Libei Liu and Sihui Li for their discussions and supports during my life in Japan.

Finally, I greatly thanks to my family for their invaluable support, hearty encouragements, and understanding.

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Chapter 2

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